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A PRELIMINARY INVESTIGATION OF AIRBORNE THORON DECAY PRODUCTS IN TWO URANIUM MINES AT ELLIOT LAKE, ONTARIO

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A PRELIMINARY INVESTIGATION OF AIRBORNE THORON DECAY PRODUCTS IN TWO URANIUM MINES AT ELLIOT LAKE, ONTARIO

by

J.L. Horwood*

SUMMARY

A gross beta method, developed to measure airborne natural radioactivity in buildings, was tested in two uranium mines near Elliot Lake, Ontario. The very large volumes of air sampled (about 4000 litres) enabled the decay of the relatively long-lived and weak activity of lead-212 (thorium-B) to be studied after the stronger activity from the daughter elements of radon had decayed away. The calculated working levels in each of this limited number of samples were comparable for the products of radon and thoron; however, this is not to suggest that the biological effects of the two species would be comparable.

A search for indications of thoron products in the results of previous measurements suggests that this species was a disturbing factor in 1976 when an Argonne Instant Working Level Meter was tested in the two mines. At that time, because all net activities were attributed to the decay of radon products, some of the values for the concentration ratios, radium A: radium B: radium C, were quite unrealistic.

This work indicates that accurate determinations of dose levels in mines in this area should include at least periodic measurements of the concentration of thoron products.

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THE INITIAL INVESTIGATION OF THE CONTRIBUTION OF THORON DECAY PRODUCTS TO RESPIRABLE DOSE LEVELS IN TWO URANIUM MINES AT ELLIOT LAKE, ONTARIO

by

J.L. Horwood

INTRODUCTION

The measurements described in this report were taken in November, 1977. Until this time the number of radioactive atoms resulting from the decay of thoron had been considered to be small relative to the number resulting from the decay of radon. However, in the experience of the author with more than two thousand spot samples, taken during the Elliot Lake Survey of Homes and in subsequent follow-up measurements in Ottawa, thoron products were frequently present and identifiable by the slow decay of lead-212 (thorium-B). A large-volume gross beta method developed by this investigator (in prep.) is sufficiently sensitive to follow and to identify the decay of lead-212, even in the lower concentrations which occur in environmental samples.

A second consideration which prompted the search for thoron products in the mine atmospheres was the well-known association of thorium along with uranium in the Elliot Lake area[1,2]. A third consideration involved calculation of the numbers of atoms per litre actually represented by the seemingly insignificant activities attributable to thoron products in the environmental samples (a summation of the counts produced from a filter during thirty or more hours of decay makes this apparent).

In the past, a theoretical consideration of the possible emanation of thoron (radon-220) from rock surfaces into the mine atmosphere suggested, on the basis of the average life of thoron gas atoms, that very little would diffuse from a rock face: thoron gas has a half-life only $\frac{1}{6000}$ that of radon (see Note 1 in the Appendix). However, another process, namely that of recoil of the product nucleus upon emission of an alpha particle from the parent, has been found to play a significant part in the liberation of radon isotopes from the surface of solids - even actinon (radon-219), the third isotope, with a half-life only $\frac{1}{80,000}$ that of radon-222 is known to reach the atmosphere in some instances[3].

When evaluating the effectiveness of the recoil process it should be noted that the parent of thoron, radium-224, emits an alpha particle whose energy of 5.68 MeV* is greater than that of the particle emitted by the parent of radon, radium-226, - energy 4.77 MeV*. Thus thoron liberated from rock by this process can originate from a somewhat greater depth and therefore volume, than can radon in the same location.

MEASUREMENTS

An Environmental Sample

Before considering measurements in the mines, it is instructive to examine an environmental sample from a test house in Elliot Lake. The decay characteristics of a combination of

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* average energy.

radon and thoron products can be followed in detail in this example using beta counts obtained every five minutes for more than ten hours. At the end of this time the combined count from lead-212 and its beta-emitting daughters (see Table 1) was still at twice the level of the background radiation; thus the decay of the sample could have been followed for many more hours.

As shown by the curve plotted in Figure 1 the initial reading at t=o, i.e. immediately after sampling, was about 18,000 counts for five minutes; of this the thoron-derived contribution extrapolated back to t=o from later readings was 360 counts for five minutes, i.e. only 2% of the total. When this deceptively small contribution is totalled throughout complete decay the total count is

$$\frac{360}{5} \times \frac{1}{\lambda} = \frac{72}{0.001086} = 66{,}000.$$

λ is the decay constant per minute of lead-212 and equals $\frac{\log \varepsilon^2}{T\ 1/2}$. T 1/2 is the half-life (in minutes here). Of this total, calculations involving the respective yields of the various beta particles together with their absorption characteristics in this (gross beta) instrument system, indicate that lead-212 accounted for 30.5%, i.e. 20,200 counts. The total radon-derived beta count from this sample was 206,000. As will be shown the proportion of the activity from lead-212 in the mine samples was several times greater.

TABLE I

U-238 SERIES (PART) WITH CORRESPONDING MEMBERS OF Th-232 SERIES (PART)

		Half-Life	α MeV	β MeV
RADIUM	Ra-226	1602 Y	4.60(6%) 4.78(95%)	
RADON*	Rn-222	3.823 D	5.49(100%)	
RADIUM-A	Po-218	3.05 M	6.00(100%)	
RADIUM-B	Pb-214	26.8 M	-	0.65(50%) 0.71(40%) 0.90(6%)
RADIUM-C	Bi-214	19.7 M	-	1.00(23%) 1.51(40%) 3.26(19%)
RADIUM-C'	Po-214	164 μS	7.69(100%)	
RADIUM-D	Pb-210	21 Y		

		Half-Life	α MeV	β MeV
THORIUM-X	Ra-224	3.64 D	5.45(6%) 5.68(94%)	
THORON*	Rn-220	55 S	6.29(100%)	
THORIUM-A	Po-216	0.15 S	6.78(100%)	
THORIUM-B	Pb-212	10.64 H	-	0.346(81%) 0.586(14%)
THORIUM-C	Bi-212	60.6 M	6.05(25%) 6.09(10%)	1.55(5%) 2.26(55%)
THORIUM-C'	Po-212	0.304 μS	8.78(100%)	
THORIUM-C"	Tl-208	3.10 M		1.28(25%) 1.52(21%) 1.80(50%)
THORIUM-D	Pb-208	(STABLE)		

NOTE: Contributions of less than 0.1% are not shown,
* only gaseous member of its group.

Samples from New Quirke

On November 2, 1977, with the full cooperation of Rio Algom Mines Ltd., it was possible to make the first *in situ* measurements using this gross beta method in a large operating uranium mine. Two air samples were collected on filters in the New Quirke Mine at the 1,935 ft. level, the first in the crusher room and the second at the station near the hoist. Beta counts were obtained from both shortly after sampling and later over a period of several hours with the same counting equipment at the Elliot Lake Research Laboratory. The results are shown in Table II and plotted in Figures 2 and 3. Note the unexpectedly low background reading underground (see Note 2 in the Appendix).

As Figures 2 and 3 indicate, the 10.65 hour half-life is again evident and unmistakable as to origin. From calibration procedures for the gross beta method described elsewhere (in prep.) total radon-derived and thoron-derived counts were calculated and will be shown subsequently in Table VII along with the next group of samples.

Samples from Quirke #1

On November 3, samples for gross beta counting were taken in the drift at level 5 of the Rio Algom Quirke #1 Mine, about five ft. downwind (west) from the portal to stope #649. Because radon concentrations are high in this location, the #1 filter, loaded by five minutes of sampling, could not be counted initially: the high count rate (estimated from the level of

TABLE II

Air Filter Samples from New Quirke Mine
Level 9, November 2, 1977

Sample #1, crusher room 3550 litres, 11:34 to 11:39h			Sample #2, near hoist 3550 litres, 11:55 to 12:00h		
Age of filter	N5(t)	(B5)	Age of filter	N5(t)	(B5)
t=1	7651*	(57)*	1	13367*	(57)*
192	1368	(88)	7	13353*	(57)*
200	1416	"	13	13000*	(57)*
267	1261	"	190	1815	(88)
277	1237	"	200	1770	"
287	1226	"	275	1476	"
471	1031	"	285	1527	"
491	999	"	460	1181	"
507	966	"	480	1216	"
520	1010	"	493	1201	"
578	893	"	505	1150	"
962	601	"	547	1114	"
1943	210	"	946	745	"
2748	94	"	1927	253	"
			2733	102	"
*counted underground					

Age of filter = minutes from end of sampling to start of count.

N5(t) = net count for 5 minutes.

B5 = background count, 5 minutes.

Parallel sampling, alpha Kusnetz, by H. Montone.

Sample #1 84 counts, after 69 minutes.

Sample #2 164 counts, after 66 minutes.

(Both Kusnetz samples were 85 litres for 5 minutes).

subsequent samples to be 500,000 counts per minute) blocked the counting system; however, the filter was useful after some hours when measured for thoron decay products. The #2 filter, from 0.5 minutes of sampling was still somewhat too active initially. Filter #3 was counted after 0.25 minutes of sampling; however, starting and stopping errors of the pump motor became relatively more significant with such a brief sampling period. The results of counting are shown in Table III and are plotted in Figure 4.

A sample was also collected for alpha counting on this occasion in stope #649 (East). The same filter was counted using both the standard Kusnetz method and also an alpha spectrometric method using a two-channel portable instrument. The latter is a prototype based on a circuit developed at the Argonne National Laboratory[4], but modified for conditions in the Elliot Lake mines and is constructed in a rugged form suitable for field testing.

The Kusnetz test established the radon* daughter concentration at 8.7 W.L. - in fact, as was shown by the spectrometer, this was a combined result of alpha particle activity from both the uranium and thorium series which did not indicate the working level resulting from the extended decay period of lead-212 and its progeny. The results from the alpha particle spectrometer are given in Table IV and are plotted in Figure 5.

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* no distinction between radon and thoron products.

TABLE III

Gross Beta Counting of Samples from Quirke #1

	Start of count t min	(1) N5 (t) (TOTAL)	(2) N5 (t) from Tn	(3)=(1)-(2) N5 (t) from Rn	(4) F (t)	(5) = (3) ÷ (4) = N5 (65)	(6) = 34 x (5)
#1	402 669 1484 1502 16000 (0)	160,000 120,700 50,200 50,000 24	(Dust) (260,000)			(840,000 estimate from #2 and #3)	(2.9×10^7)
#2	64 389 654 1469 (0)	110,100 17,000 13,000 5,470	24,670 (26,445)	85,430	1.022	83,600	2.8×10^6
#3	6 12 376 643 648 1456 (0)	123,700 117,000 8,730 6,759 6,626 2,740	13,300 13,200 (13,360)	110,400 103,800	2.289 2.192	48,200 47,400	1.64×10^6 1.61×10^6

For Sample #1: total (estimated) radon-derived count = 2.9×10^7
total " thoron-derived count = 4.8×10^7

N5(t) = net count 5 minutes at time t; F(t) = a decay factor = N5(t) ÷ N5(65).

TABLE IV

Alpha Counts from Filter Obtained in Stope #649
(Quirke #1, November 3, 1977)

t → minutes	0	3.5	17.5	65	242.5	1392.5
Counting → Interval		1-6	15-20		238-247	1388-1397
*counts/min →						
RaA + ThC(+)		4201	923		132	40.1
RaC' + ThC(-)		7974	7512		275	64.1
TOTAL			8435		407	104
ThC + ThC'			463 ←	440	363 ←	104
RaA + RaC'			7972 →	3760 →	407 68 339	
ThC + ThC' (Average)	457			426 3760	(339+363)½ 351	
TOTAL ALPHA	(KUSNETZ)			4185		
RaA + RaC'	(RELATIVE)		2.12	1.000	0.0180	1.45×10^{-15}
Pb-212	1.000	0.9962	0.9812	0.9318	0.7685	0.2204

* arrows indicate order of calculations

$$\text{Pb-212 (at t=o)} = \frac{351}{0.7685} \times \frac{T_B}{T_B + T_C} = 417$$

(T_B and T_C are half-lives of ThB and ThC).

Because there was little or no ventilation of this stope, the radon products (RnP) and the thoron products (TnP) may be assumed to be close to equilibrium – this will be substantiated later in this report by earlier measurements with the Argonne Instant Working Level Meter in 1976. Once collected on filters in this location the TnP will quickly reach <u>transient</u> equilibrium[5,6]; then the observed activity of the daughters will be somewhat greater than that of their parent because their decay lags that of the parent on the filter. The total number of lead-212 atoms (equilibrium condition) collected from 34 litres, i.e. lead-212 plus progeny equals 417, (the lead-212 count rate at t=o), X $\frac{1}{\varepsilon}$ X $\frac{1}{\lambda_\beta}$

ε = the counting efficiency (compared with Kusnetz = 0.31) = 0.15;

λ_β = the decay constant for lead-212 = 0.001086 (per minute).

$\therefore$ No. of lead-212 atoms per litre =

$$417 \times \frac{1}{0.15} \times \frac{1}{0.001086} \times \frac{1}{34} = 7.53 \times 10^4.$$

Because the mine atmosphere could be assumed in this case to be in equilibrium, the numbers of atoms of the various thoron products were proportional to their half-lives; and the number of (ThC + ThC') atoms $= \frac{7.53 \times 10^4 \times 60.6}{10.64 \times 60} = 7.1 \times 10^3$ per litre.

The total number of atoms, each of which can contribute an <u>average</u> potential alpha decay energy of 7.8 MeV (see Note 3, Appendix)

$= 7.53 \times 10^4 + 7.1 \times 10^3 = 8.24 \times 10^4$.

$\therefore$ W.L.T. (working level due to TnP)

$$= \frac{8.24 \times 10^4 \times 7.8}{1.30 \times 10^5} = 4.9.$$

Using the count ratio, $\frac{C(RnP)}{C(Total)} = \frac{3760}{4185}$, calculated for the alpha spectrometer in Table IV at t = 65 min, and applying this to the Kusnetz determination (energy response of the two alpha counters assumed to be similar):

$$W.L.R. = \frac{3760}{4185} \times 8.7 = 7.8$$

(W.L.R. and W.L.T. are not combined, because much of the ingested Pb-212 will not be retained long enough for decay to occur).

Calculation of Working Levels from the Beta Measurements

This calculation has been postponed until this point in the report because it is much more complex than for the alpha measurement:

(1) the gross beta method has been calibrated previously for radon decay products by means of parallel sampling with Kusnetz equipment (Gourgon, Priv. Com.). However, in the case of thoron decay products no data from parallel sampling are yet available for the beta method, therefore the calibration factor must be calculated in relation to RnP and will involve a small decrease (~12%) in efficiency because of greater absorption of the lower energy beta particles from lead-212 in the doubly-folded filter;

(2) the state of equilibrium of the mine atmosphere in the case of the samples from New Quirke is not known precisely, but must be estimated from the degree of ventilation and from the decay characteristics observed with one of the two samples. The effect on the dose level is not large (10%, or less) but should be considered because of the varied conditions to be expected throughout an operating uranium mine;

(3) the variation in response (TnP to RnP) of the gross beta equipment to gamma emitters (see Table I) collected on the filter has not been calculated. However, measurements of background counts suggest that the efficiency of the "pancake" Geiger-Meuller tubes in regard to gamma radiation will be less than that of the beta detector (a plastic scintillator) in the Argonne Instant Working Level Meter; i.e. less than 10%. Thus the assumption that the additional count from gamma emission in the case of the TnP is in the same proportion to that added to the RnP, will not produce a significant error;

(4) differing effects from conversion electrons produced by radon daughters and thoron daughters will be small because the particles are of low energy and will be removed to a large degree by absorption in the doubly-folded filter and in the mica window of the Geiger-Meuller tubes[7,8].

A first step in calculating the relative beta efficiencies, RnP to TnP, is to consider the overall efficiency, ε, to be the product of two components; $\varepsilon = E_x \cdot E_R$

where E_x represents the transmission efficiency, i.e. the fraction

of the emitted ß particles which penetrate the folded (#541) filter and the mica window of the GM tube; and E_R represents the remaining factors involved in the total efficiency. E_R includes: filter retention efficiency (when calculating concentrations in the mine atmosphere), transmission efficiency of the bronze screen protecting the mica windows of the GM tubes, sample geometry as related to relative areas of the portion of the filter measured by the detectors compared with the total active area of the filter, edge effects, sample/detector distance - all contributing to the difference from 4π sample geometry. The values of these factors, measured and/or calculated will be included in a separate report describing the gross beta method (in prep.). The value of E_R (TnP) is taken here to be very close to that of E_R (RnP). E_x, however, has a different value for the combined products of each series - as well as for the components of each - so that E_x varies as transient equilibrium develops on the filter.

Calculation of the transmitted fraction of each of the fifteen (continuous) beta particle energies involved in this work was based on a relationship given by Evans based on an empirical mass absorption curve for the natural beta emitters resulting from the work of Curie et al[9,10]:

$$\text{i.e. } \rho\chi = 41\, E_m^{1.14}\ g/cm^2$$

$$\text{and } I = I_o\, \ell^{-\left(\frac{17}{E_m^{1.14}} \cdot \rho\chi\right)}$$

where ρ is the density of the absorber in g/cm^3,

χ is the thickness in cm,

E_m is the maximum energy in MeV of the continuous energy distribution of the ß emission, I is the transmitted intensity and I_o is the incident intensity.

For the doubly-folded filter plus the mica window (4mg/cm^2) of the GM tube, the four possible mass absorption paths ($\rho\chi$) are: 9.4, 10.6, 24.0 and 29.8 mg/cm^2. The transmitted fraction was calculated for each path and the combined result for each beta energy group is given Table V.

The table shows that the decay of 100 atoms of Pb-212 will eventually produce 190 beta particles, of which 121 can penetrate the absorbers; i.e. the transmission is $\frac{121}{190} = 0.64$ for thoron products. Similarly the decay of 100 atoms of Pb-214 through Bi-214 will produce 196 beta particles of which 140 can penetrate for detection; the transmission is $\frac{140}{196} = 0.71$ for radon products. (The latter value was found experimentally to be 0.70).

Another step in transforming alpha and beta count rates into "working level" values is the consideration of disequilibrium among the decay products, both in the mine atmosphere and on the filter paper. Because Quirke #1 is not an operating mine, there is little ventilation in the region where the samples were taken, therefore, the filters from that mine were considered to be in transient equilibrium at all stages of counting. In the case of New Quirke, the ventilation was strong in the crusher room and at the nearby station. The sample taken at the station was counted <u>in situ</u> at 1, 7 and 13 minutes after the end of sampling with resulting evidence of a large excess of radium-A; the decay of this excess on the filter (by alpha emission only) delayed the apparent decay of the subsequent beta emitters compared with the characteristic decay rate experienced in the case of moderate ventilation in buildings.

TABLE V

Transmission and Combined Yield of Beta Emitters in Radon and Thoron Progeny

Nuclide	Maximum β Energy, MeV	Fraction Emitted	Fraction Transmitted	Net Fraction after Transmission	Combined Yield
Pb-212	0.346	0.81	0.388	0.314	0.395
(Th-B)	0.586	0.14	0.580	0.081	
Bi-212	1.55	0.05	0.828	0.041	0.528
(Th-C)	2.26	0.55	0.886	0.487	
Tl-208	1.28	0.25 } 0.96*	0.790	0.198	0.287*
(Th-C")	1.52	0.21 } X 0.36	0.825	0.173	
	1.80	0.50 } = 0.35	0.851	0.426	
	(* 36% abundance, 0.797 X 0.36 = 0.287)				
	TOTAL	1.90			1.21
Pb-214	0.65	0.50	0.616	0.308	0.610
(Ra-B)	0.71	0.40	0.645	0.258	
	0.98	0.06	0.734	0.044	
Bi-214	0.4	0.09	0.4	0.036	0.788
(Ra-C)	1.00	0.23	0.739	0.170	
	1.51	0.40	0.824	0.330	
	1.88	0.09	0.85	0.077	
	3.26	0.19	0.922	0.175	
	TOTAL	1.96			1.398

The growth of lead-212 (thorium-B) and its daughters toward transient equilibrium is shown graphically in Figure 6. Note that the total alpha curve rises less steeply from zero in its initial stages because no alpha particle emitters are present in the decay chain below lead-212 until some of the latter has decayed. The change in slope of this curve may be utilized to determine the "age" of the mine air with respect to thoron daughters (understood to mean "after lead-212"). In such measurements, because the alpha particles from polonium-218 and bismuth-212 have almost identical energies and therefore cannot be resolved, the 8.78 MeV alpha particle from polonium-212, one that is very easily resolved, must be used for this purpose. Two counting intervals, each of 5 to 10 minutes (because the activities are low particularly under conditions of strong ventilation), separated by an interval of about 20 minutes, are tentatively suggested.

In contrast to the total alpha curve in Figure 6, it should be noted that the total beta curve rises quickly from zero in its initial stages. This is because lead-212 is produced very shortly, on the average, after thoron leaves the rock. As shown in Table I the half-lives of thoron and its daughter are very short, so that 99% of thoron converts to lead-212 within six minutes. In the case of the gross beta equipment used here, significant filtering of the soft beta component from lead-212 causes the beta curve to start effectively near a value of 30%. With other beta systems the effect of internal absorption in the sample and in the window of the detector may be less. In the case of the latter systems,

such as the Argonne IWL Meters having only a single thickness of filter membrane interposed between the active layer of the sample and the beta detector, it is expected that the beta curve would start at about 40%.

In assessing the effect of the proportionality between lead-212 and its daughters in the mine atmosphere, it will be found that complete removal of the daughters, never achieved in practice, will only reduce the respirable dose levels by $\frac{T_C}{T_\beta + T_C}$ where T_β is the half-life of thorium-B, i.e. 10.64 hours, and T_C is the half-life of thorium-C, i.e. 1.01 hours. Note that the population of thorium-C' (polonium-212) atoms in the mine atmosphere is at all times negligible (under conditions of equilibrium among the thoron daughters with a combined dose level of lead-212 and daughters of 0.33 W.L.T., it would be necessary <u>on average</u> to sample at 17 litres per minute continuously for more than nine years to collect a single atom from the atmosphere). Thus it may seem that an accurate determination of the state of equilibrium among the thoron daughters is not essential for a satisfactory measurement of the dose level; however, if thoron-derived activity is measured at say four hours after sampling and the resulting activity is extrapolated back to the time at which the radon daughters were measured, the thoron-derived contribution, calculated in this way for subtraction from the total count, will be too great under conditions of extreme disequilibrium. Fortunately, the thoron-derived activity is generally much less than that of the radon products which in itself will limit the error in apportionment of activity.

Working in the opposite direction, the error could be more serious; that is, if thorium-C' activity were measured only once shortly after sampling, it could be difficult to determine the true lead-212 concentration, unless the ratio of the radium-A to radium-C' count rates could serve as a suitable measure of equilibrium conditions that would be applicable to thoron products as well. The validity of this assumption can be tested by counting again after four hours when the radon derived activity has decayed to a negligible level and lead-212 with its daughters is known to have reached transient equilibrium.

In applying the previously defined relationship $\varepsilon = E_X . E_R$ (page 12) to radon decay products, RnP values are developed from parallel sampling side by side with the alpha Kusnetz method (Gourgon, Priv, Com), of activity on a filter during sampling and the subsequent decay during counting[11,12]. These values, for five minutes sampling and five minutes counting, are as follows:

$$W.L.R. = \frac{N5\ (t)}{190.1 \times L \times F\ (t)} .$$

where: N5 (t) is the net 5-minute count starting t minutes after the end of sampling;

L is the rate of flow in litres per minute;

F (t) is a decay factor which becomes 1 at t=65, the constant, 190.1, was experimentally determined and represents the combined result of parallel sampling over a range of concentrations in eleven different locations (Gourgon, Priv., Com.).

The calculated number of beta disintegrations occurring between 65 and 70 minutes after the end of five minutes of sampling:

(Ra-B + Ra-C) = 3667 before absorption,

and = 2680* after absorption,

(per litre per minute for W.L.R. = 1).

From the above:

$\varepsilon_{RnP}/E_x.E_R = 190.1 \div 3667 = 0.0709,$

$\varepsilon_{RnP}=E_x.E_R = 190.1 \div 2680 = 0.0518,$

and $E_x = 0.0518 \div 0.0709 = 0.731,$

(or 2680 ÷ 3667).

To determine ε_{TnP} for thoron products during "later" counting (i.e. after say four hours when transient equilibrium has been established on the filter), in transient equilibrium the ratios of the activities Th-B, Th-C and Th-C" are 1, 1.105 and 1.110[13]. Table V is used to provide the following:

	Fraction Emitted	Combined Yield Trans. Equm
Th-B	0.95 X 1.00 = 0.95	0.395 X 1.00 = 0.395
Th-C	0.60 X 1.05 = 0.633	0.528 X 1.105 = 0.583
Th-C"	0.35 X 1.110 = 0.388	0.287 X 1.110 = 0.319
	Total = 2.001,	and 1.297.

E_x (for TnP) = 1.297 ÷ 2.001 = 0.648,

and ε_{TnP} = 0.65 X 0.0709 = 0.0461.

Note: E_x will be slightly lower for early counting of samples out of equilibrium: proportion of Pb-212 will be greater.

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* the calculated value for t=o to t=∞ is 91,000;
91,000 ÷ 2680 = 34.0 (used in column 6 of Table III).

In Table VI the equations used to develop Table V and Figure 6 were employed to calculate the contribution from thoron products for each of the samples listed. The result from a very low level sample (7.5×10^{-4} c/min/l) collected in Room 122, 555 Booth Street has been included to illustrate the usefulness of large-volume sampling in situations where the Kusnetz method (with the usual 85-100 litre sample) may be unreliable. The ages of the air listed are estimates which are assumed to be reasonable; if necessary, the decay characteristic of a sample (near t=o) can be used in gross beta counting to suggest the age of the air as is shown in Figure 7.

Table VI shows that the age of the air has little effect on the total working level of the thoron products; however, under conditions of very strong ventilation, the total beta count (from thoron products) may be reduced below 60% of the extrapolated transient equilibrium value at t=o. This may have a significant effect on the residual count attributed to radon products in some circumstances where thorium levels are high, such as at Agnew Lake. The change in the working level ratio, TnP to RnP, should also be noted. This change will be referred to later in the discussion.

Indications of thoron products in measurements prior to November 1977

Although thoron products make up a significant part of the total dose in each of the mine samples reported here, the number of samples is small; many more measurements will be required to determine "average conditions" in the mines at Elliot Lake.

TABLE VI

Calculated working level values, RnP and TnP, for samples listed

	#1 New Quirke	#2 New Quirke	#1 Quirke 1	#2 Quirke 1	#3 Quirke 1	Test House Elliot Lake	Elliot Lake Research Lab.	555 Booth St.
Location	1935 Ft CRUSHING R.	1935 Ft STATION	- DRIFT,	LEVEL 5 NEAR STOPE	#649 -	BASEMENT	BASEMENT	ROOM-122
Date	- 2/11/77 -		-	3/11/77	-	4/11/76	3/11/77	4/11/76
Flow Rate cuft/min	26	25	27	27	26	30	27	62
Sample volume liters	3690	3550	3830	380	~185	4260	3830	35,000
Count/5 min at t=o (1)	1680	2060	260,000	26,400	13,400	360	115	26.4
Count/min/liter	9.11×10^{-2}	1.16×10^{-1}	13.6	13.8	~14.5	1.69×10^{-2}	6.0×10^{-3}	7.5×10^{-4}
LEAD-212 c/min/l	2.78×10^{-2}	3.54×10^{-2}	7.9×10^{4}	8.1×10^{3}	4.1×10^{3}	1.10×10^{2}	35	8.1
(Thorium-B) atoms/l	25.6	32.6	3800	3900	~4000	4.8	1.7	~0.3(2)
Working Level	0.033	0.042	5.0	5.0	~ 5.3	0.0062	0.0022	~0.0004
BISMUTH-212 Age of air min	16(3)	22	- -	(OLD)	- -	40	(OLD)	-
(Thorium-C) % Equilibrium	17	22	- -	~100	- -	37		-
Working Level	0.0005	0.0009	0.48			0.0002	0.0002	-
(TOTAL) WORKING LEVEL TnP	0.034	0.043	5.5			0.0064	0.0024	~0.0004
RADON DAUGHTERS C-65 (4)	3250	5600	8.4×10^{5}	8.4×10^{4}	4.8×10^{4}	5680	2130	
Working Level RnP	0.023	0.042	5.8			0.035	0.015	
W.L. Ratio TnP:RnP	1.48	1.02	0.95	(0.63)		0.18	0.16	

(≈ Stope #649E 4.9 ÷ 7.8 = 0.63)

(1) Extrapolated back from subsequent readings,
(2) Faster filter used, estimated correction applied.
(3) Reasonable age assumed to illustrate effect on results.
(4) Count/5 min. normalized to 65 min.

In addition to follow-up surveys of K.C. Cheng of the Elliot Lake Research Laboratory and D.W. Carson[14] of this laboratory who are studying the prevalence of thoron products, it has been possible to make use of measurements taken with an Argonne Instant Working Level Meter at Elliot Lake in 1976[15].

A re-examination of the measurements made at that time now indicates that readings of apparent relative (activity) concentrations of radium-A, radium-B and radium-C which seemed anomalous, or even impossible to explain in terms of radon products alone can now be explained satisfactorily by a partial contribution from the decay products of thoron. A Rolle type diagram[16] is an effective method of demonstrating the unusual nature of the original determinations.* An interpretation has been made based on several possible factors, including the presence of lead-212 and its daughters; all of the factors could affect the positions of readings plotted on this diagram. The readings were taken in August and September 1976, while the Argonne instrument was on loan at Elliot Lake through the courtesy of the US Bureau of Mines and the Argonne National Laboratory.

In Figure 8, a Rolle diagram has been plotted showing examples of radium-A, -B and -C, values obtained from the literature; they represent a range of both mine, air and environmental samples. Each point represents two ratios: the apparent, measured

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* The author is indebted to J.R. Johnson, C.N.R.L., Chalk River, Ontario, for pointing out the usefulness of this type of diagram in a pre-print of his report[19].

activity ratio, radium-B to radium-A, plotted as the abscissa and the ratio radium-C to radium-A as the ordinate. Points plotted farther away from the origin at 0,0 toward P(1,1) indicate a lower degree of ventilation; also those closer to the 45° line, denoted by M, show a mixing of low-activity air with a proportion of air tending toward a condition of equilibrium, whereas the points closer to the curve denoted by N show a mixing of low-activity air with "young" air, i.e. higher activity concentrations of both radium-A and radium-B than of radium-C.

It has been shown that the area bounded by the curves M and N represents all possible (activity) concentrations of radon decay products[16,19]. Exceptions to the latter would include: (1) factors relating to the sample, including disturbance by the act of measurement and the presence of another radioactive species; (2) factors relating to the measuring instrument, including mis-adjustment, drift due to low battery voltage and temperature effects.

In Figure 9 points, plotted from measurements taken with Argonne IWLM at Elliot Lake in 1976, are significantly displaced from the area bounded by M and N. At first sight, it may be tempting to attribute this displacement to the presence of various combinations of lead-212 and its decay products in the samples; however, a careful analysis of the constants stored in the computational section of the instrument (A.N.L., IWLM Unit #3) indicates that factors, additional to the presence of lead-212 and products, have contributed substantially to the positions of the points in Figure 9. These factors are related to the stability and adjustment of the instrument.

To verify the presence of thoron, as indicated by lead-212 and its products and to separate its influence from other factors of an instrumental origin and to evaluate the effect of the other factors, the following procedures were adopted:

(1) the more detailed results from the IWLM when operated in the "calibrate mode" were examined (e.g. #3 and #4 in Figure 9) because accumulated counts were given in each of the three channels, radium-A and radium-C' (alpha) and radium-B + radium-C (beta), for the time periods 0-2, 0-5, 0-30, 0-35 and 39-41 minutes;

(2) the well-known exponential equations defining the build-up of activity on a filter during sampling (in this case during two minutes) were evaluated to provide the number of disintegrations per minute at t=o (the end of sampling) for 100 pci/l atmospheric concentrations, each and separately, for radium-A, -B and -C[11,12]. The resulting values were then applied to a further set of exponential equations used to define the decay of various proportions of the radon daughters during decay and measurement on the filter[20]. These results were integrated for the time intervals specified in (1) above. Then it was possible to determine E_x and E_β, the efficiencies of the alpha and beta channels of the IWLM (Unit #3) and to show that the measured rates of decay, alpha and beta, were indeed slower than those predicted for radon daughters, thus providing an indication, independent of the displacements in Figure 9, that thoron daughters were detectable from the measurements. In some of the calculations, the values in the A and C' channels were added and considered as "total alpha" to avoid partitioning or overlap errors between the channels that could have resulted from drift, misadjustment, low battery voltage and/or poor resolution;

(3) a programmable calculator was loaded (Commodore PR-100, 10-memories and 67 program steps used) with the matrix given in Note 4 of the Appendix. This enabled concentrations of radium-A, -B and -C to be determined in one operation from various count yields chosen for the 0-2 min counting interval of the instrument. (Note that the constants are specific to the particular instrument, IWLM Unit #3, and will vary somewhat for other A.N.L., IWLM,s;

(4) in addition to facilitate the reverse process, the inverse matrix was calculated. Thus, it was possible to move computationally from a chosen position on a Rolle diagram to counts proportional to concentration (not absolute unless the "working level" value is also known). This matrix* is given in Note 5 of the Appendix.

Rates of decay determined from samples measured in the "calibrate mode" are summarized in Table VII by a set of ratios: the net count per minute in each of the separate time intervals (0-2, 2-5, 5-30, 30-35 and 39-41 minutes) was divided by the net count per minute over the full interval, 0-35 minutes. Thus the counts were normalized so that they could be compared with the count rates in the intervals calculated from theory for radon daughters with no contribution from thoron products. The ratios of "measured" to "calculated" results are plotted in Figures 10 to 13.

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* Because all coefficients are non-zero in this matrix and the PR-100 calculator is limited to 71 program steps, the final five steps were hand-entered to solve all three equations with a single program.

TABLE VII

Decay Ratios of IWLM Samples Divided by Radon Products

Time Interval Minutes		Radon Products (Calculated)	Sample #1 (5 Aug.)	Sample #2 (5 Aug.)	Sample #3 (6 Aug.)	Sample #4 (6 Aug.)
0-2	α	1.00	0.978	0.965	0.971	0.942
	β	1.00	0.976	0.981	0.982	0.959
2-5	α	"	0.984	0.972	0.961	0.964
	β	"	0.976	0.978	0.974	0.977
5-30	α	"	1.002	1.004	1.010	1.010
	β	"	1.002	1.001	1.001	1.004
30-35	α	"	1.025	1.034	1.004	1.026
	β	"	1.026	1.024	1.029	1.020
39-41	α	"	0.990	1.036	1.010	1.016
	β	"	1.040	1.044	1.031	1.049

The increased slope of a line from the horizontal is an indication of an increased relative contribution from thoron products. However, the slopes should not be interpreted too rigorously at this stage because:

(1) Ra-A, -B and -C activity ratios of 1:1:1 were assumed for the calculated reference count yields (not an unreasonable assumption for the degree of ventilation in this location);

(2) runs #1 and #2 were made August 5, and runs #3 and #4 were made August 6; some adjustment to the circuit controls was made between those days.

It may be possible at some stage to interpret different slopes (same sample) of lines derived from alpha counting versus slopes from beta counting as an equilibrium factor for the thoron products; it is less likely in Figures 10 to 13 because of the low degree of ventilation. However, in the case of each line, the difference in slope from the horizontal indicates a modification of the natural decay rate of radon products alone toward a more gradual combined decay rate resulting from the additional (slower decay) contribution of lead-212 and its daughters (half-life, 10.64 hours).

The displacements in Figure 9 of points outside the area defined by MN can now be examined in more detail. The use of the two matrices permits "changes" to be made to the instrument and/or mine atmosphere in a theoretical or computational manner in order to study the resulting calculated relative apparent concentrations of Ra-A, -B and -C, as depicted on a Rolle diagram; the inverse

process is also possible. Changes made and shown in Figure 14 include the following:

L-1 displacement along this line away from P, or along another parallel line is caused by the addition of TnP (or a decrease in RnP) in nearly equilibrium proportions. Disequilibrium, i.e. air "younger" in regard to TnP, causes a modification of the slope toward L-1A and then L-1B;

L-2 displacement along this line, or along a parallel line, is caused by a change in the proportions of the total alpha counts directed into the respective A and C' channels. The count yield ratio, A:C', as measured for TnP does not differ greatly from the ratio for RnP, also in general the count yield RnP >> TnP, thus thoron products are not involved in this displacement. Contributing factors of a more likely nature are an incorrect point of division between the A and C' alpha channels resulting from misadjustment, or a shift in the alpha pulse distribution caused by temperature, low battery voltage, increased absorption on the filter from heavy dust and/or humidity, deterioration of the alpha detector;

L-3 displacement along (or parallel to) this line results from a change in the measured (B+C) count while at the same time conditions remain fixed in the alpha detection system. Because of the high degree of sensitivity to changes in gain in the beta detection system as evidenced by the scale on L-3, it is important to stabilize the photomultiplier tube against temperature and voltage changes and to calibrate the IWLM instrument at a representative ambient temperature;

L-4 displacement along (or parallel to) this line results from a change in the A count only, such as from readjustment of the threshold control. The change can also result from natural changes in the measured system such as "plating out". Note the slope of L-4 is very nearly 45°, which may be interpreted as an equal effect on the B/A and C/A ratios.

The apparent B/A and C/A ratios were modified in a "mechanical" way in the case of one measurement, indicated "S" in Figure 9, by the addition of a 100-mesh screen over the input to the filter to collect the unattached ions, mainly radium-A, thus increasing the apparent relative concentrations of radium-B and radium-C. Displacement of the plotted position of this sample from other members of its group is evident in Figure 9.

Comparisons of rates of decay and displacements on a Rolle diagram as illustrated by the lines, L-1, etc., will be applied to samples, #3 and #4, taken August 6, 1976, in Quirke #1. The calculated count rate ratios of pure RnP (for Ra-A, -B, -C = 1:1:1) in the intervals 0-5 to 0-35 minutes are:

$$\underline{\text{Alpha}}: \quad \frac{C\ 0\text{-}5}{C\ 0\text{-}35} = 1.463, \text{ and}$$

$$\underline{\text{Beta}}: \quad \frac{C\ 0\text{-}5}{C\ 0\text{-}35} = 1.215$$

The ratios obtained for the samples (i.e. of counts per minute) are:

#3 $\underline{\text{Alpha}}: \quad \frac{C\ 0\text{-}5}{C\ 0\text{-}35} = \frac{27,132}{5} \cdot \frac{35}{134,491} = 1.412$

$\underline{\text{Beta}}: \quad = \frac{56416}{5} \cdot \frac{35}{332,445} = 1.188$

#4 Alpha: $= \frac{22664}{5} \cdot \frac{35}{113690} = 1.395$

Beta: $= \frac{48396}{5} \cdot \frac{35}{287510} = 1.178$

Each ratio is lower than the corresponding calculated value, thus suggesting that the samples are not radioactive as a result of radon products only; i.e. $\frac{C\ 0\text{-}5\ + T_1}{C\ 0\text{-}35 + T_2}$, could be used to express the ratio, where T represents TnP. For equilibrium conditions, $T_1 \approx T_2$ except for a small correction for the rate of decay of lead-212. Making the latter correction and solving for the equivalent T in the interval 0-2 minutes, produces for #3, T alpha = 891 and T beta = 2640; and for #4, T alpha = 1001 and T beta = 3120. T_β is approximately 3 T_α as would be expected. There are two beta particles emitted for each alpha particle emitted in this part of the thorium series (see Table V) and the measured counting efficiencies are: $E_\beta = 0.3$ and $E_\alpha = 0.2$, (E includes a small contribution from gamma radiation; thus $\frac{T_\beta}{T_\alpha} \approx 2 \times \frac{0.3}{0.2} = 3$. The total TnP alpha contribution, T_α, is divided between the A and C' channels in the ratio, previously mentioned, of 0.38 to 0.62. Thus the TnP contributions assigned to each channel in the IWLM become:

	$\frac{A}{339}$	$\frac{B + C}{2640}$	$\frac{C'}{552}$,
and #4	380	3094	621.

The effect of subtracting these values from the respective count yields for the samples will be tested using a Rolle diagram as shown in Figure 15. The activity ratios as originally measured by the IWLM are plotted as points P_{30} and P_{40}. After subtracting

the above values for the TnP and solving the matrix, the activity ratios (for RnP only) are plotted as points P_{31} and P_{41} respectively.

If drift in the alpha pulse amplifier, or a small misadjustment of the point of division between the A and C' channels, is an explanation of the residual displacement from the point P(1,1), this can be corrected in the calculations by a shift in the L-2 direction by transferring counts from the C' to A channels. In this way, moving on the diagram to points, P_{32} and P_{42}, 403 counts = 3.58% of the total alpha was transferred from C' to A for sample #3 and 159 counts = 1.77% for #4. Possibly an average value (2.7%) would be more representative of the actual requirement. It should be noted that these percentage values are related to the steepness of the alpha pulse amplitude continuum and do not necessarily represent changes in amplifier gain of that magnitude.

The effect of random fluctuations in the count rates on the activity ratios depicted here, is suggested by the length of the line, P_{42} to P_{43}. The square root of the count in the B+C channel was subtracted from the beta count; i.e. $\sqrt{16212}$ = 127 counts. Points, P_{32} and P_{42}, are thus within "statistical range" of P(1,1). The latter point may not quite represent the actual activity ratios for the RnP fraction: the B/A and C/A ratios may be somewhat less than 1.00. The above steps were taken to illustrate that a relatively simple combination of factors can explain anomalous positions on a Rolle diagram.

The values used for the alpha-emitting thoron products in samples #3 and #4 are consistent with the count in the same location measured by CANALPH-3 in November, 1977. In the latter,

the count rate 0-2 minutes, was calculated from alpha counts taken after several hours; this result is consistent with the 1976 values:

Comparing TnP count rates (alpha) in Stope #649 East

Count (0-2 min) per minute/litre for E_α = 1, 3 November, 1977;
(from Table IV) ≡ 457 X 9.9989 ÷ 34 ÷ 9.15 = 90 c/min/l,
#3 ≡ 891 ÷ 2 ÷ 22.4 ÷ 0.213 = 93 c/min/l, 6 August, 1976;
#4 ≡ 1001÷ 2 ÷ 22.4 ÷ 0.213 = 105 c/min/l, 6 August, 1976.

A Rolle diagram will now be used to examine other sets of results obtained with the Argonne IWLM in 1976. These are plotted in Figure 16. The points designated P_{50} and P_{60} are average values from two groups totalling 29 samples and shown individually in Figure 9. The two groups of samples, collected in or near stope #649 on the same day, can be shifted into the area MN by subtracting similar proportions of thoron products; i.e. by moving the same distance in the L-1 direction to the points P_{60-61} suggests a higher level of ventilation, in which case the slope of the shift will change toward the direction P_{60} to P_{62}.

In the remaining group of samples in Figure 16, numbered (5) to (9), there is evidence of stronger ventilation, of a much higher relative contribution by thoron products, of significant disequilibrium among both radon and thoron products and of a progressive change in concentration of both groups of products during the total sampling period. This becomes more apparent when the counts for each sample in the A, B + C and C' channels are calculated using the inverse program. The results are given

in Table VIII. The minimum Ra-A contribution is estimated by subtracting 0.6 X count in C' channel from the A channel. Thus Ra-A, which tends to represent the radon concentration, increases during the interval. At the same time, the counts in the B + C and C' channels decrease, indicating a decreasing thoron level. The fact that the B + C channel decreases by about 1/3 at the same time as the A channel increases significantly, shows that the thoron-derived activity is at least comparable with that derived from radon. This suggests that the TnP working level may well be much higher than the RnP working level at this degree of ventilation and if so, would help to confirm the higher TnP:RnP ratios in New Quirke as summarized in the last line of Table VI.

DISCUSSION AND CONCLUSIONS

The evidence of significant dose levels of thoron products in mine atmospheres at Elliot Lake contained in the earlier data obtained with the Argonne Instant Working Level Meter supports the findings of November 1977; however, only a systematic and extensive survey can determine whether the measurements reported here represent something close to average conditions.

In assessing factors which tend to produce higher concentrations of the decay products of one radioactive species compared with those of another, it should be remembered that rate of decay can influence concentration levels both upward and downward. In the case of radon, its relatively slow decay provides time for it

TABLE VIII

Samples from 5 level drift, about 30 feet east (upwind) of the portal to stope #650, 1/9/76.

DESCRIPTION	CONCENTRATIONS OR COUNTS		
SAMPLE { APPARENT RnP CONC. pCi/l	RaA	RaB	RaC
SAMPLE { COUNTS/2 min from INV. PGRM	(A, α)	(B + C, β)	(C', α)
#5, 11:40 AM, WL* = 0.061	4.92	11.7	0.85
	(71)	(307)	(30)
(A) - 0.6 (C') = Minimum RnP	53		
#6, 12:00 PM, WL = 0.045	5.48	8.26	0.61
	(77)	(218)	(21)
	65		
#7, 12:20 PM, WL = 0.046	5.64	8.47	0.62
	(80)	(224)	(22)
	67		
#8, 12:40 PM, WL = 0.043	5.39	7.77	0.51
	(76)	(203)	(19)
	65		
#9, 1:00 PM, WL = 0.046	7.17	7.76	0.26
	(100)	(195)	(15)
	91		

* IWLM READING

to find its way around barriers, enables transport in water with subsequent emanation and allows buildup and subsequent withdrawal from fissures and broken rock by changes in atmospheric pressure, that is by "pumping", whereas the very rapid decay of thoron requires direct injection into the mine atmosphere if the decay products are to become air-borne. However, once the atmosphere has been reached the situation is reversed. If a given air mass requires say one hour to move along the ventilation circuit of a large mine before leaving the mine, during that interval only 0.75% of the radon travelling in that air mass will have decayed to form hazardous products. Thoron, on the other hand, decays so rapidly that it is impossible to remove any significant fraction of it by any conceivable level of ventilation. Thus, it is really lead-212 that effectively is removed by the ventilation system. However, at this stage, thoron has already transformed into a solid product and of this more than 6% will decay to become a potential alpha emitter during this period of one hour.

The fact that thoron and its products may in some ways behave quite differently from radon and its products makes it important that research results obtained in relation to the latter group receive independent appraisal. Examples which come to mind include:

(1) the use of membrane barriers, epoxy paints, etc., may be much more effective in preventing thoron entering the mine atmosphere. There is not time for gaseous thoron to find its way around such a barrier. In fact, merely wetting a rock surface could be effective if the layer of water is thick enough to prevent penetration by the

recoil product nuclei. Wetting could be tested in an area which is adequately isolated from any thoron products carried along a mine ventilation circuit;

(2) it would be interesting to determine the build up of TnP relative to RnP along a mine ventilation circuit, and whether the buildup is greater in dry areas. There is a suggestion in Figure 9 that the relative proportion of thoron products may be somewhat lower farther into stope #649 west than in the not-so-wet area near the portal. Could thoron products have entered the stope from the drift rather than having been produced there?;

(3) thoron product to radon product ratios should be investigated at all levels of ventilation to verify the indication (supported by the findings of Kerr) that the TnP:RnP ratio becomes greater at higher levels of ventilation (Priv. Com);

(4) the fact thoron cannot be stored in fissures and piles of broken rock, for later "pumping" by air pressure changes, makes it important to study the range of variation of the daughters at fixed locations. If the range is found to be small, this could reduce the frequency of monitoring. Also, it might enable some simplification of monitoring instruments and procedures;

(5) because thoron travels a relatively short distance during its average lifetime of seconds, it should be possible to locate regions of production with a sampling probe designed to admit the gas only. Resulting charged ions of polonium-216 (thorium-A) could be collected electrostatically within the probe and identified and counted using alpha spectrometry;

(6) because of its relatively long half-life of 10.64 hours, with subsequent emission of alpha particles by its daughters, lead-212 could serve as an excellent, natural, radiotracer in studies of bronchio/lung retention times of aerosols. Sputum samples properly prepared for high-resolution low-background alpha spectrometry could provide confirmatory data for comparison with results calculated using bronchio/lung models.

Additional surveys may show that the levels of thoron products are at times camparable with those of radon in these mines; however, the calculations of Jacobi and others, which took into account the much slower decay of the thoron products, suggest that there is not an increased hazzard as a result of these levels (21). Although the discovery of thoron products could bring added complexity to the more sophisticated health monitoring procedures (IWLM's, etc), this can be offset by the capabilities of modern electronic instrumentation.

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D.W. Carson, J.V. Krzyzewski and J.F. Tippins of the Mineral Sciences Laboratories provided very useful technical assistance as follows:

(D.W. Carson)

- operation of the Argonne I.W.L.M. during calibration and numerous tests in homes and underground;
- construction of the detector heads for the gross beta method;
- operation of gross beta equipment (calibration and survey) in several hundred Elliot Lake homes;
- construction of a modified A.N.L. 2-channel portable alpha spectrometer; obtaining of the initial mine sample with this spectrometer in stope #649, November 3, 1977.

(J.V. Krzyzewski)

- provision of the (pancake type) Geiger-Meuller tubes and of counting equipment for the gross beta method.

(J.F. Tippins)

- construction of the two lead shields for the gross beta method.

/tab

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APPENDIX

Migration of Radon and Thoron into a Mine Atmosphere

Note 1

Lambert and Bristeau demonstrated by simple radioactive measurements "that the diffusion of radon is very low within various monocrystals and that it cannot explain the emanating power of rocks"(22). They concluded that the only mechanism necessary to explain the loss of radon from the rock in which it formed was recoil, and that the loss by this process occurred from a layer within 500 ängstroms of the surface. Their value for the diffusion coefficient of radon at ordinary temperatures is 10^{-21} to 10^{-22} cm^2 sec^{-1}.

Tanner in his review of the migration of radon gives a value for the diffusion coefficient of lead in zircon and monozite of 10^{-21} to 10^{-22} cm^2 sec^{-1}(23). The latter value was obtained by Nicolaysen(24).

Using the value which favours most the release, i.e., $D = 10^{-21}$ cm^2 sec^{-1}, in the one-dimensional diffusion equation clearly demonstrates that diffusion is quite ineffective in the release of radon from within a rock crystal:

$$C = Co \exp(-\sqrt{\lambda/D}\, Z) \text{ where}$$

C = concentration at a distance Z

Co concentration at Z = 0

λ is the disintegration constant which

= 2.10×10^{-6} sec^{-1} for radon and

= 1.27×10^{-2} sec^{-1} for thoron.

D is the diffusion coefficient.

$\sqrt{D/\lambda}$ is defined as the diffusion length; the gas concentration at this distance is 0.37 of the initial concentration Co

For λ's = (Rn) and D = 10^{-21} cm^2 sec^{-1} the following values are obtained for the thicknesses, Z, shown:

TABLE IX

Diffusion of Radon within a Rock Crystal

Z in cm	$3X10^{-7}$	10^{-6}	$3X10^{-6}$	10^{-5}
ängstroms	30	100	300	1000
microns		0.01	0.03	0.1
C/Co for radon	$1.1X10^{-6}$	$1.25X10^{-20}$	$1.97X10^{-60}$	∿0

For Z = 1 ängstrom, C/Co for thoron is $3.3X10^{-16}$ under these conditions.

Kigoshi used the rate of ejection of alpha-recoil thorium-234 from zircon sand to determine the alpha-recoil range, 550 ängstroms,(25). The rate of supply, Q, of the Th-234 atoms from the solid surface is given by: $Q = \frac{1}{4} L S \rho U \lambda u$ where L is the α-recoil range of Th-234 in the silicate, S and ρ are, respectively, the surface area and density of the silicate, u is the number of U-238 atoms in 1 g of silicate, and λu is the decay constant of U-238.

Because the number of atoms of radon and thoron released from rock is proportional to the area of the surface, it is reasonable to consider finely-divided rock which will have a relatively large surface area relative to its volume. With finely-divided material such as sand and mud, the gases must

still diffuse from the porous medium, containing air, water or both, into the mine atmosphere. Although diffusion alone may not cause escape to occur within a reasonable time relative to the half-life, escape may well be assisted by transport mechanisms such as the movement of air, water and (in the case of crushing, blasting, etc.) the movement of rock - it may be a combination of these mechanisms.

Table X shows the extent to which radon and thoron can penetrate various typical media as a result of diffusion (alone). The values were calculated from diffusion coefficients quoted by Tanner (23). The table should assist the investigator, with a knowledge of conditions underground, to determine the more likely regions of radon and thoron emanation. It should also assist in designing isolated tests of emanation from various types of radioactive material, and in the design of barriers which should be particularly effective in the case of thoron.

TABLE X

The Diffusion of Radon and Thoron through Various Media

Porous Medium D in cm^2 sec^{-1}	for	C/Co for Diffusion Distances, Z, in cm						
		0.1 cm	0.3	1	3	10	30	100
Building Sand (Air + 4% moisture) $D = 5.4X10^{-2}$	Rn	1.00	1.00	0.99	0.98	0.94	0.83	0.54
	Tn	0.95	0.86	0.62	0.23	$7.8X10^{-3}$	$4.8X10^{-7}$	$8.7X10^{-22}$
Fine Sand, mainly quartz (Dry) $6.8X10^{-2}$	Rn	1.00	1.00	0.99	0.98	0.95	0.85	0.57
	Tn	0.96	0.88	0.65	0.27	0.013	$2.3X10^{-4}$	$1.7X10^{-19}$
Fine Sand (Air + 15% moisture) $1X10^{-2}$	Rn	1.00	1.00	0.99	0.96	0.87	0.65	0.23
	Tn	0.89	0.71	0.32	0.034	$1.3X10^{-5}$	$2.1X10^{-15}$	$1.1X10^{-49}$
Fine Sand (Air + 17% moisture) $5X10^{-3}$	Rn	1.00	0.99	0.98	0.94	0.81	0.54	0.13
	Tn	0.85	0.62	0.20	0.0084	$1.2X10^{-7}$	$1.7X10^{-21}$	$6.1X10^{-70}$
Mud (Air + 37% moisture) $5.7X10^{-6}$	Rn	0.94	0.83	0.54	0.16	0.0023	$1.2X10^{-8}$	$4.4X10^{-27}$
	Tn	0.0089	$7.1X10^{-7}$	$3.2X10^{-21}$	-	-	-	-
Mud (Air + 86% moisture) $2.2X10^{-6}$	Rn	0.91	0.75	0.38	0.053	$5.7X10^{-5}$	$1.9X10^{-13}$	-
	Tn	0.00050	$1.3X10^{-10}$	-	-	-	-	-
Water $1.13X10^{-5}$	Rn	0.96	0.88	0.65	0.27	0.013	$2.4X10^{-6}$	-
	Tn	0.035	$4.3X10^{-5}$	$2.8X10^{-15}$	-	-	-	-
Air $1.2X10^{-1}$	Rn	1.00	1.00	1.00	0.99	0.96	0.88	0.66
	Tn	0.97	0.91	0.72	0.38	0.039	$5.8X10^{-5}$	$7.4X10^{-15}$

APPENDIX

Note 2 — Background count underground

Underground the background count from the two pancake-shaped Geiger-Meuller tubes was unexpectedly low. Compared with a count on the surface which was consistently about 88 counts for the usual 5-minute counting period (longer periods up to 1000 minutes or more were used and the value for 5 minutes calculated from the result), the count underground was 57 counts for 5 minutes. This is attributed to the shielding against the effects of cosmic radiation provided by several hundred feet of rock.

An average thickness of two inches of lead was used around the GM tubes with the expectation that this thickness might be somewhat too low because of gamma radiation from the surrounding radioactive rock. The results indicate that for detectors such as these at least, much ligher shielding could be used to aid portability: 0.5 to 1 inch thickness should be adequate.

Note 3 — Average energy of the alpha particles from ThC and ThC'

Referring to Table I:

for every 100 atoms of ThC which decay,

25 decay with an energy of 6.05 MeV = 151 MeV
10 decay " " " " 6.09 " = 61 "
and the remainder after β decay to ThC'
65 decay with an energy of 8.78 MeV = 571 "
Total 783 MeV

i.e. 7.8 MeV average energy per particle.

Note 4

Coefficients for IWLM #3

Using counts of A, B+C, C' to solve for concentrations (pCi/ℓ):

pCi/ℓ	A	(B + C)	C'
RaA	0.0735	0	-0.0103
RaB	-0.00309	0.0495	-0.110
RaC	0.000085	- 0.003566	0.0654

V: 24 litres (12 ℓ/min.)

Note 5

Inverse Matrix for IWLM #3

Using concentrations of RaA, -B, -C to solve for counts arising from A, B+C, C^1:

Concentrations, pCi/ℓ (coefficients for)

Counts	RaA	RaB	RaC
A	13.61	0.1756	2.436
B+C	0.9212	23.00	36.68
C'	0.03255	1.2544	17.401

TABLE XI

Reference No.	Company Name	U_3O_8 %	U_3O_8 Extraction by Leaching %	ThO_2 %	ThO_2 Extraction by Leaching %	Leaching Test Work SR Report No.	Mineralogy SR Report No.	Main Ion Exchange SR Report No.
5/55-17	Algom Nordic	0.16	94	0.02	92	359/55	259/54	376/55
4/57-6	Algom Nordic	0.13	94	0.028	89			
4/57-6	Algom Quirke	0.089	80	0.05	68			
11/54-2	Bicroft - Croft	0.08	96	0.057		344/55	303/55	360/55
12/54-4	-Center Lake	0.11		0.10			305/55	
10/55-6	Black Bay	0.13	94			391/56		
12/55-6	Blue Rock	0.13	87	0.05		406/56	402/56	
3/56-16	Can. Met.	0.10	93	0.03		429/56	429/56	
1/56-22	Cavendish	0.23	93	0.29		409/56	410/56	
11/55-9	Cayzor	0.80	98	0.002		392/56		
1/56-20	Can Dyno	0.095	95	0.01	50	414/56	377/55	430/56
5/56-24	Con Denison	0.23	96	0.07	83	439/56	438/56	451/56
	Eldorado Beaverlodge	0.24	92*	0.005				
	" Port Radium	0.66	96	0.003				
11/55-2	Faraday	0.12	96	0.09	81	394/56	395/56	411/56
1/56-23	Greyhawk	0.081	94	0.08		401/56	404/56	
4/56-4	Gulch	0.088	95	0.01		426/56	427/56	437/56
8/53-13	Gunnar	0.18	94			212/54		201/53
12/55-7	Halo	0.19	95	0.06		406/56	413/56	
6/55-9	Lake Cinch	0.17	95			366/55	364/55	
12/55-11	Lake Nordic	0.071	90	0.03				
9/55-4	Lorado	0.26	95	0.002		375/55	381/55	390/56
6/56-10	Millikin Lake	0.095	93	0.04		445/56	440/56	
4/56-10	Nesbit Labine	0.22	98			436/56	284/54	
2/56-3	Panel	0.11	90	0.06		431/56	420/56	
12/53-8	Pitch Ore	0.048	67*			225/54	222/54	
2/54-9	Pronto	0.094	89	0.05		249/54	218/54	276/54
5/57-24	Pronto	0.14	89	0.032	25			
9/56-12	Rare Earth	0.11		0.14				
8/54-4	Rayrock	0.43	98	0.002		447/56	455/56	457/56
11/56-8	Rexspar	0.10	92	0.13		475/57	465/57	476/57
7/53-15	Rix	0.28	96			172/53	181/53	
6/55-12	Stanleigh	0.071	96	0.03		442/56	441/56	
7/56-16	Stanrock	0.10	95	0.07		452/56		
12/55-10	Spanish American	0.11	89	0.05				

* Carbonate leach

Note: Typical Uranium Ore values

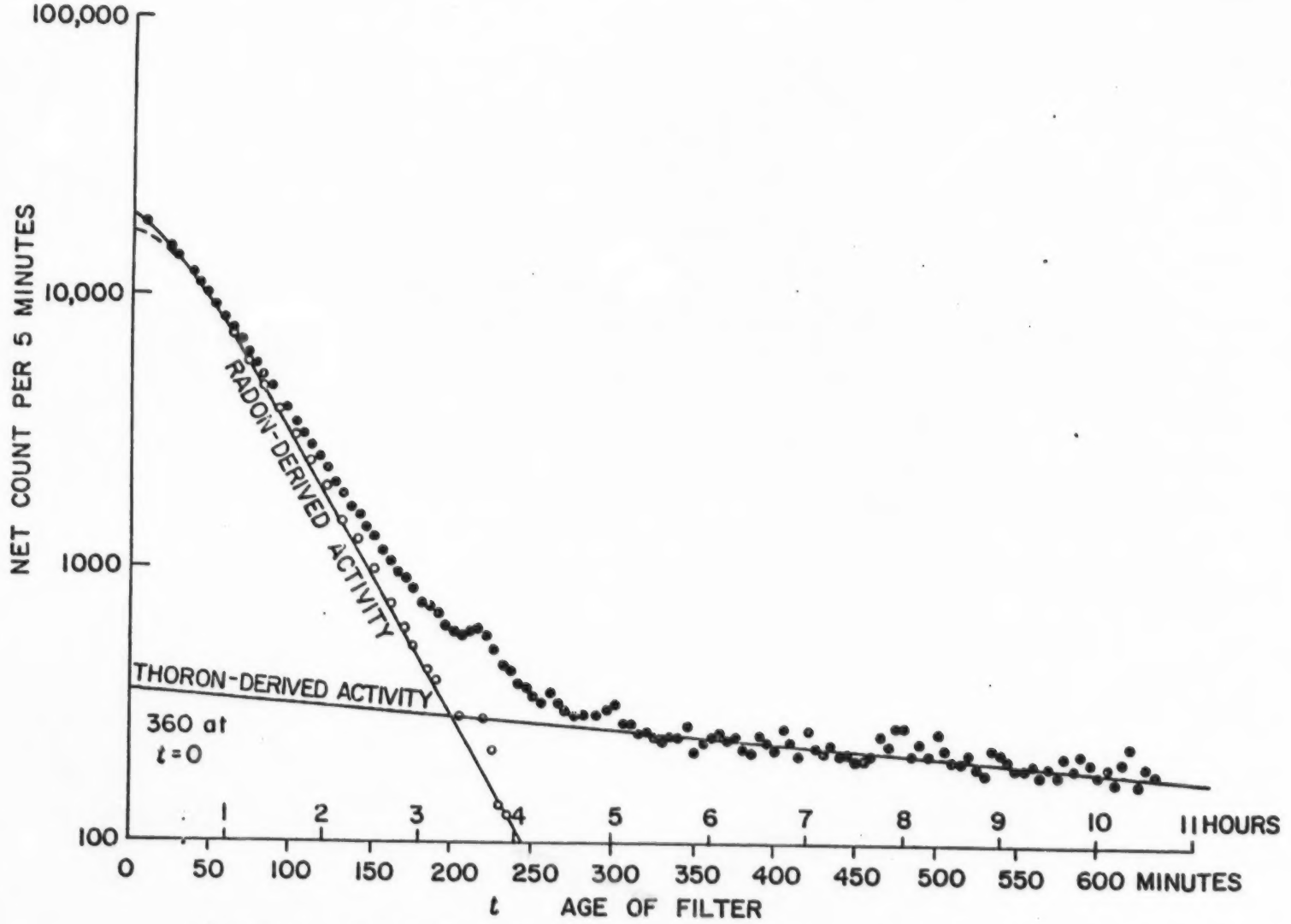

Figure 1. Environmental Sample from Test House at Elliot Lake

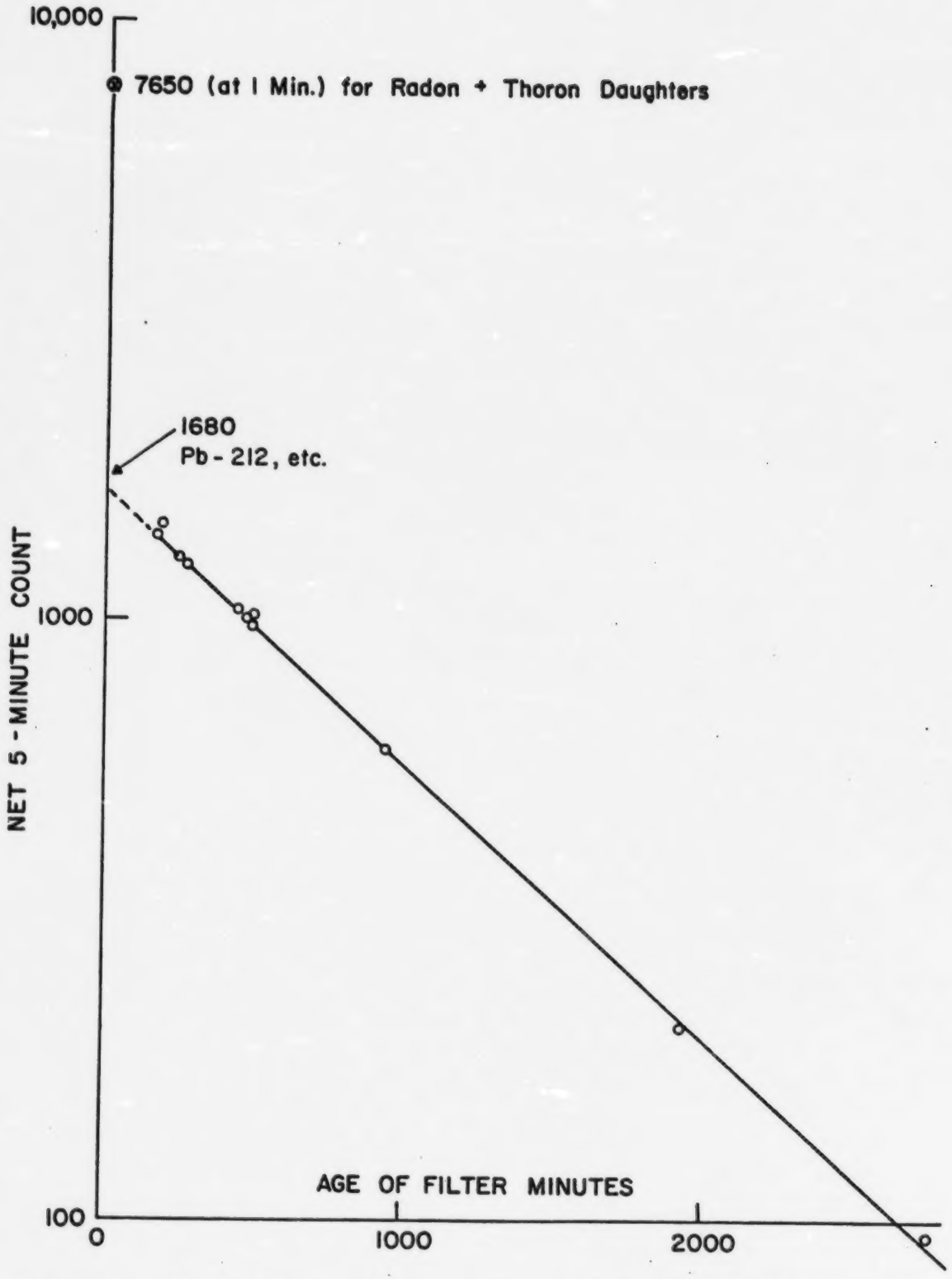

Figure 2. Sample #1, Crushing Room, New Quirke, 2 November, 1977

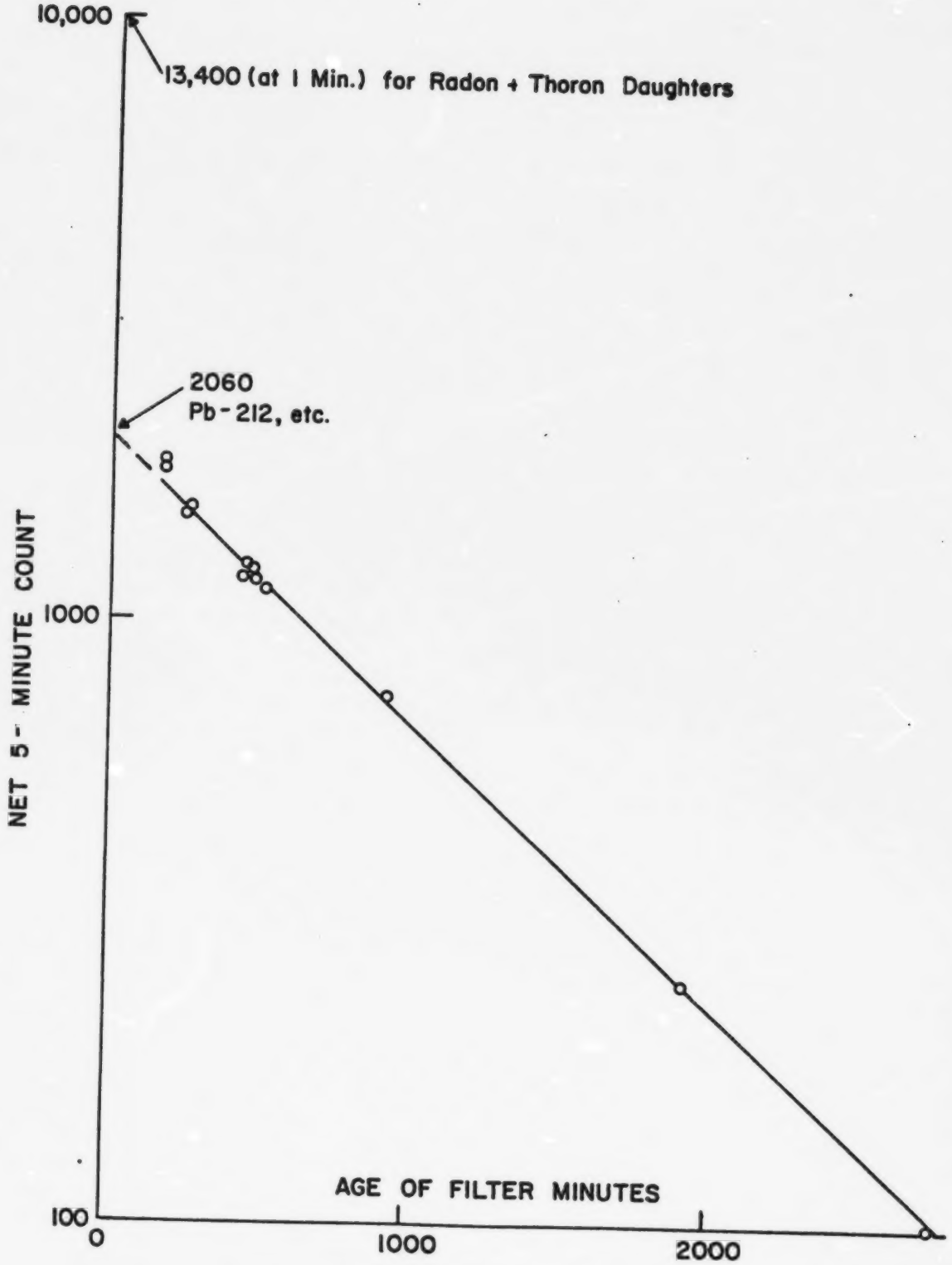

Figure 3. Sample #2, Near Hoist, New Quirke, 2 November, 1977

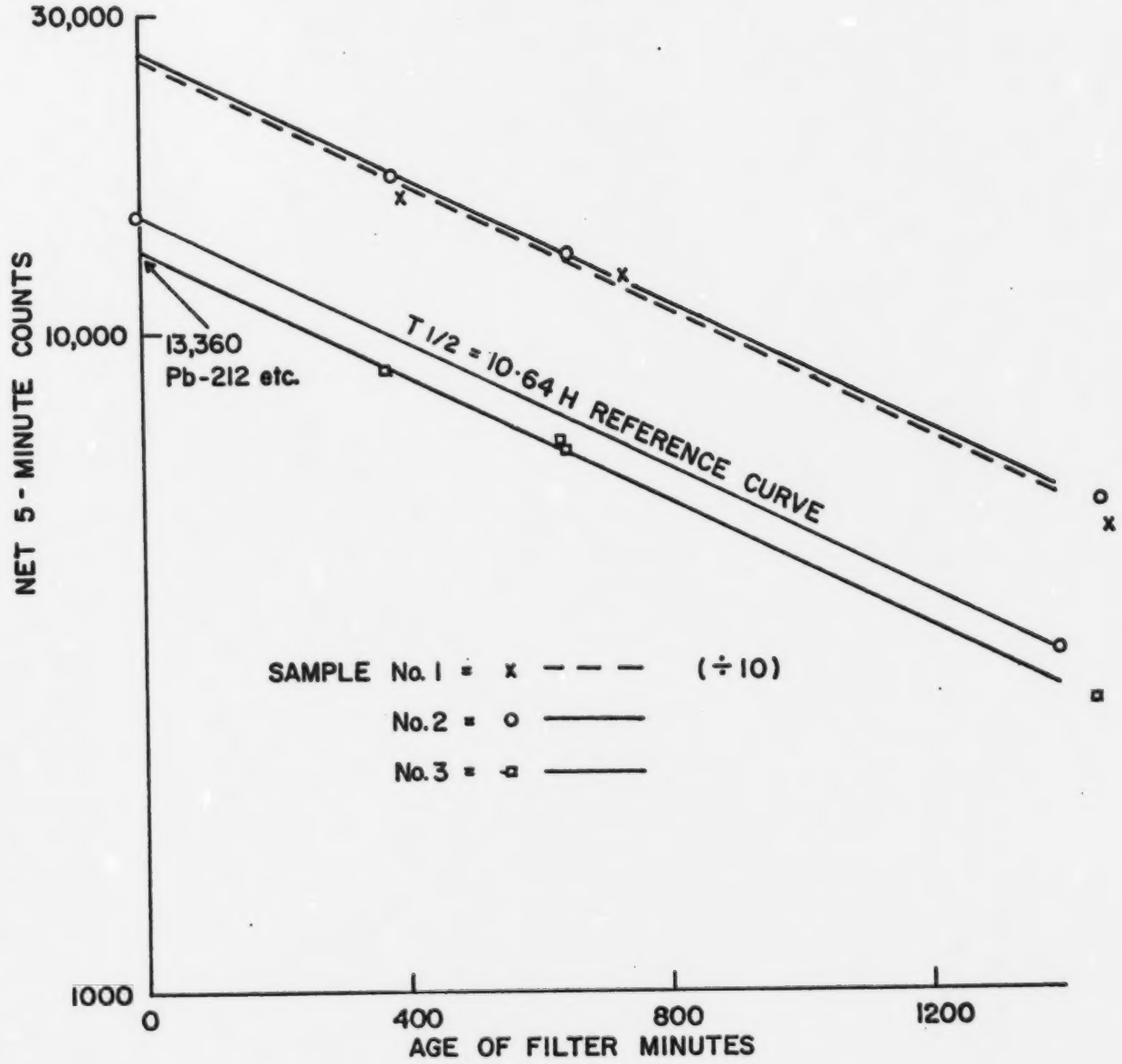

Figure 4. Samples #1, 2, 3 in Drift near Stope #649, Quirke #1, 3 November, 1977

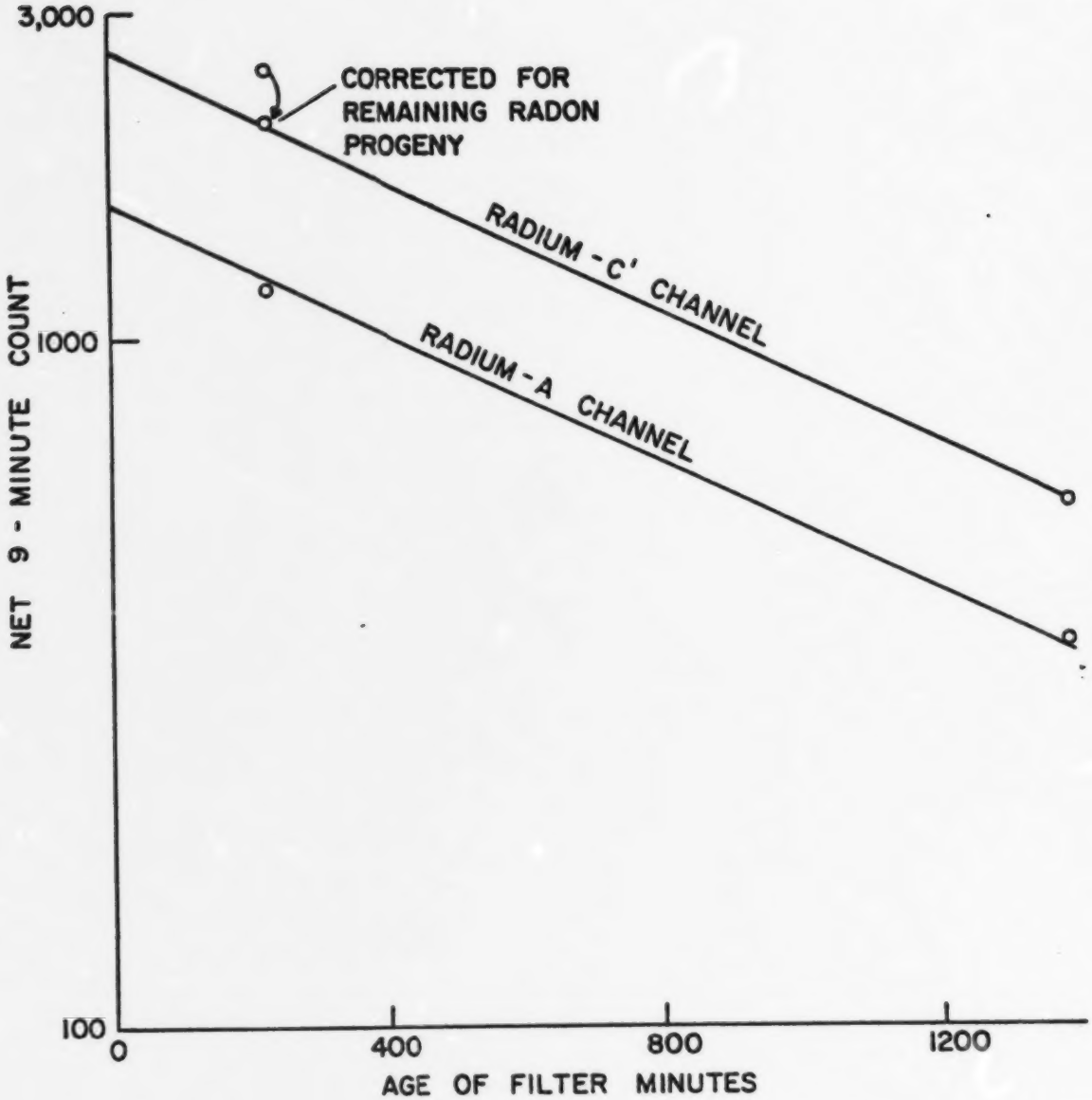

Figure 5. Sample #4, Stope #649, Quirke #1, 3 November, 1977

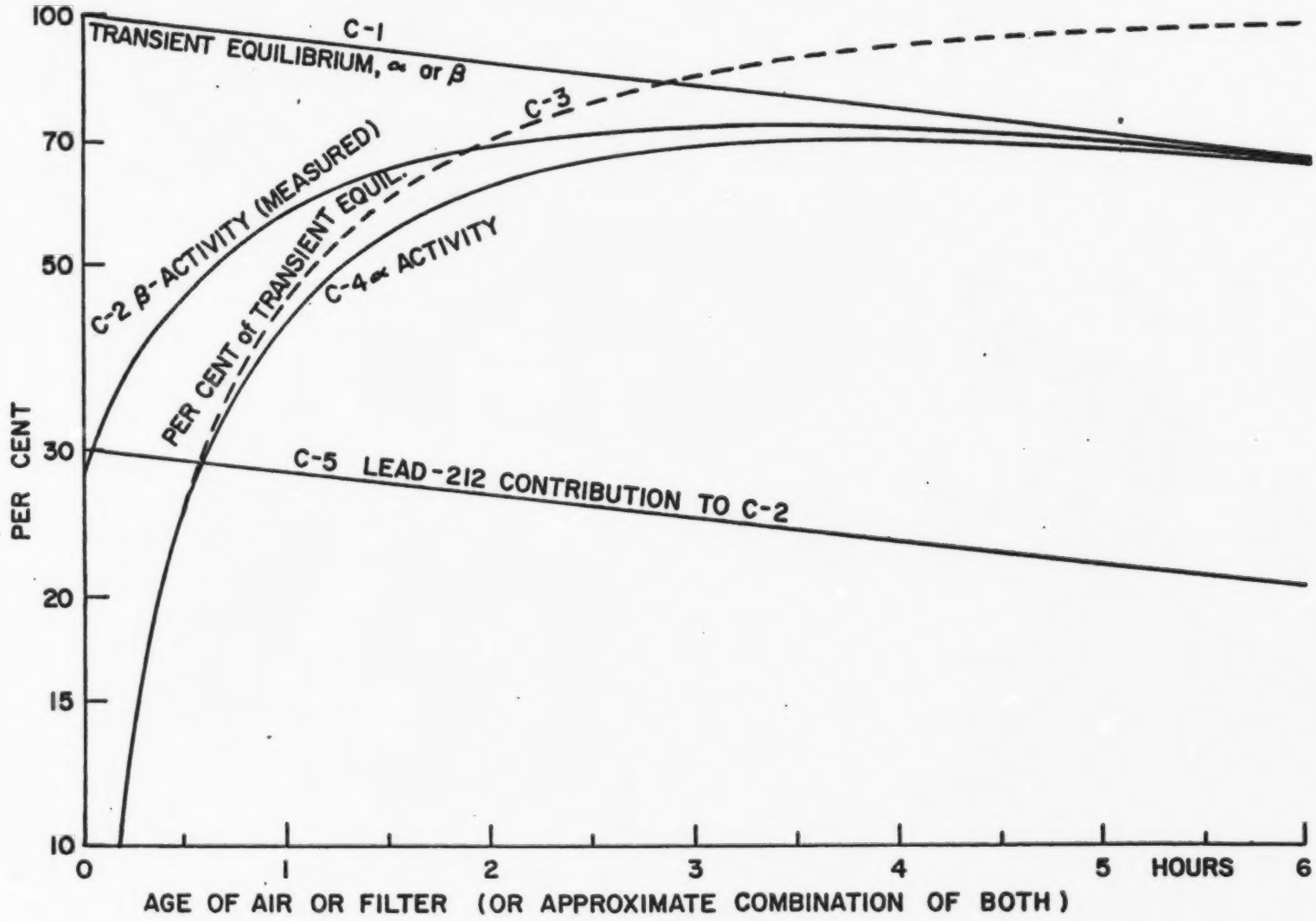

Figure 6. Growth of Lead-212 and Daughters toward Transient Equilibrium

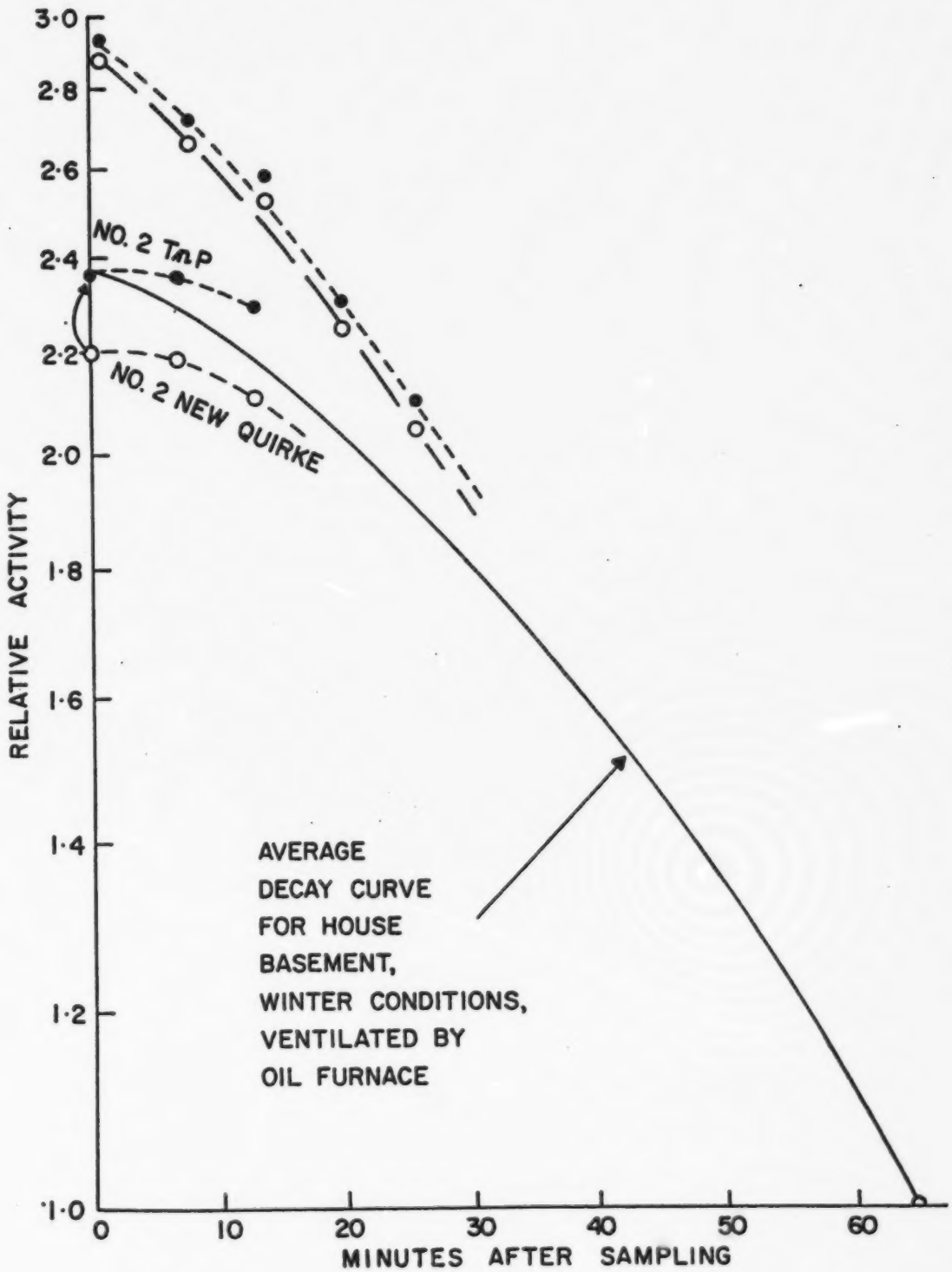

Figure 7. Effect of the Degree of Ventilation on the Decay of Gross Beta Samples

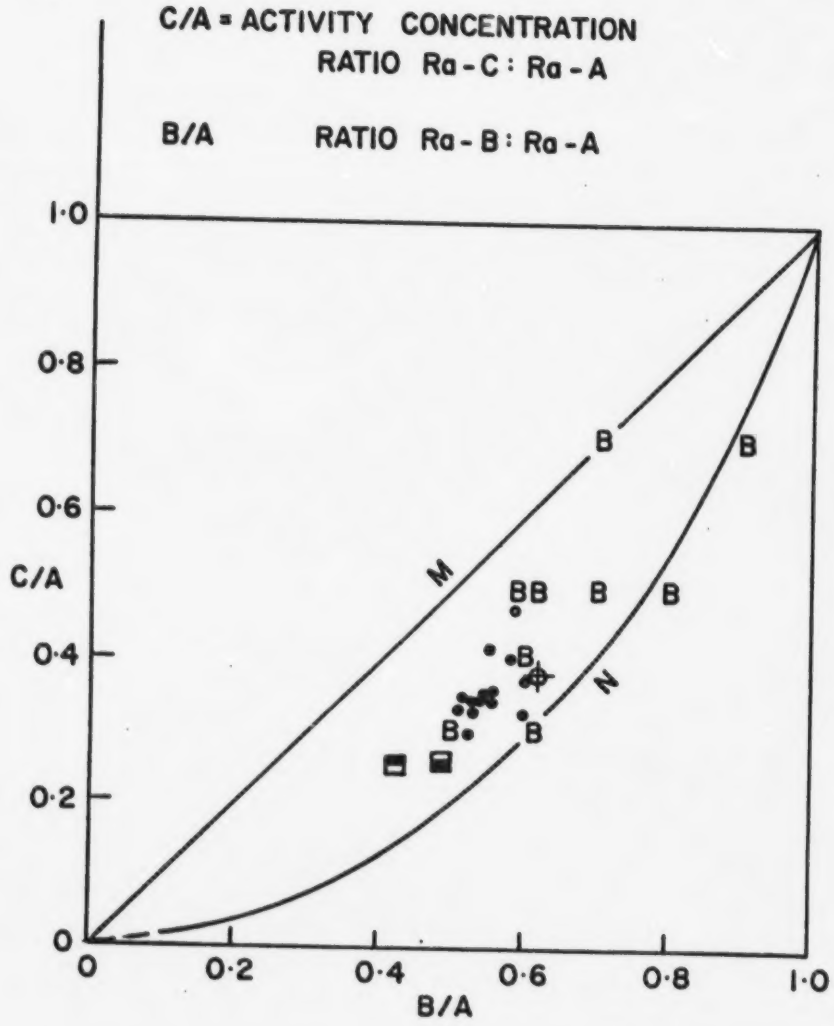

Figure 8. Rolle Diagram of Ra-C/Ra-A and Ra-B/Ra-A Ratios from Values in the Literature

(11) MARTZ, et al.

• Uranium Mine
Mean Working Level ($\overline{WL}$) = 6.96

(17) GEORGE

⌖ Sidewalk New York City
$\overline{WL}$ = 0.0007, (8 readings)

⊙ Sterling Forest (50 Mi N of NYC)
$\overline{WL}$ = 0.0009 (6 readings)

▣ Basement, H.A.S.L., NYC
$\overline{WL}$ = 0.023 (11 readings)

▣ 5th Floor, H.A.S.L., NYC
$\overline{WL}$ = 0.001 (11 readings)

(18) GEORGE & BRESLIN

B Uranium Mine, Beaverlodge, Sask.
$\overline{WL}$ 0.3 to 11 (Ventilation Nil to 8000 CFM)

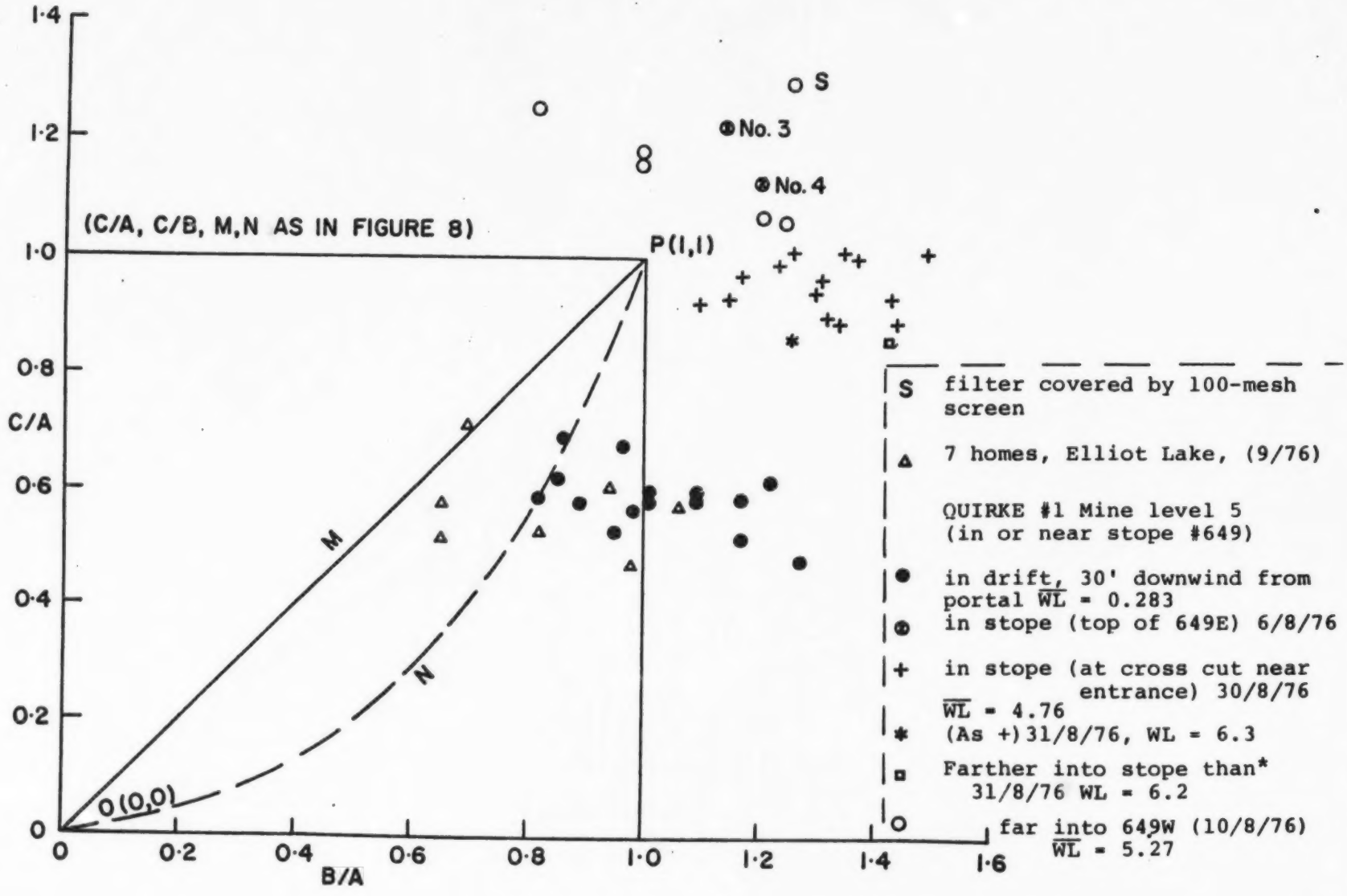

Figure 9. Rolle Diagram from Measurements with Argonne IWLM, Unit #3 (1976)

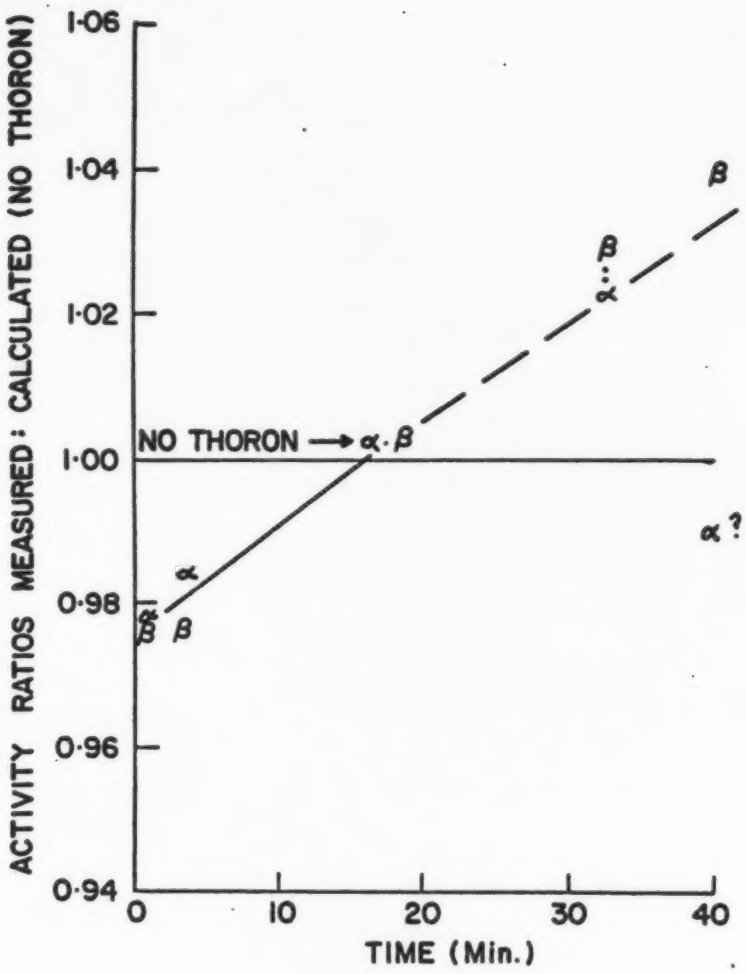

Figure 10. Sample #1, Table VII, Ratios Measured to Calculated Values (calculated = 1.00, represented by horizontal line)

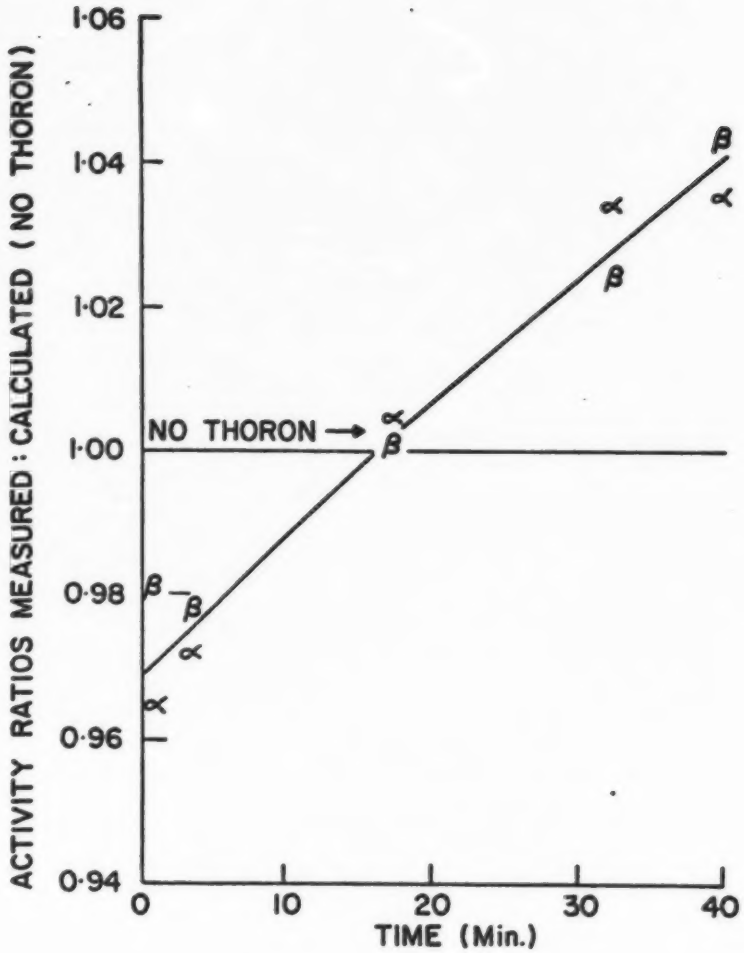

Figure 11. Sample #2, Table VII, Ratios Measured:Calculated

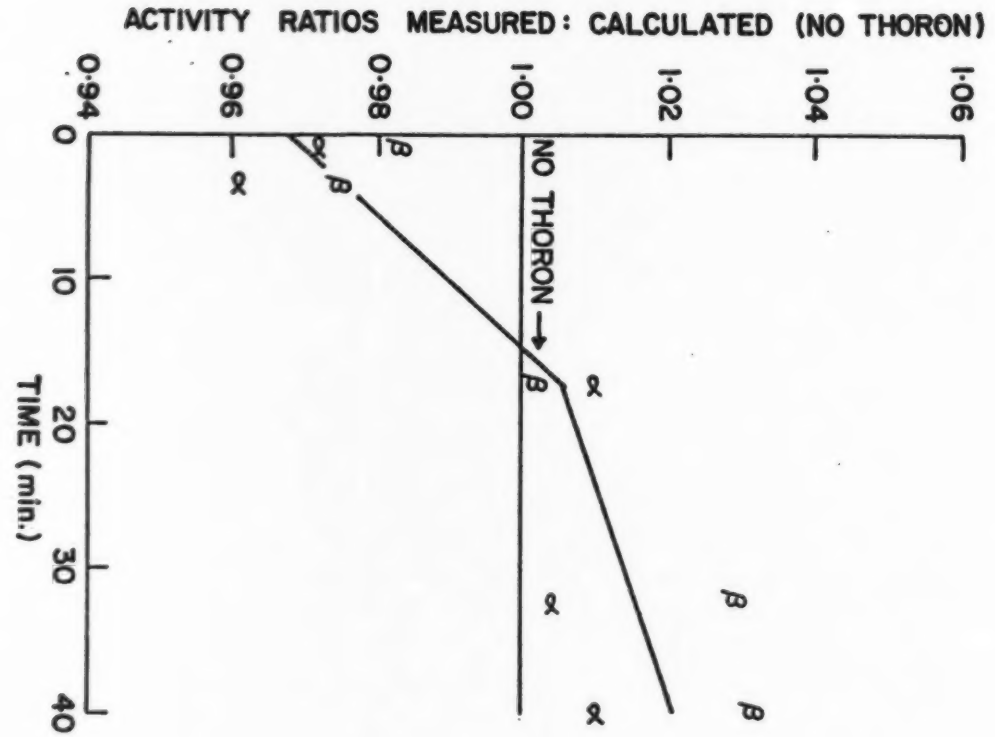

Figure 12. Sample #3, Table VII, Ratios Measured:Calculated

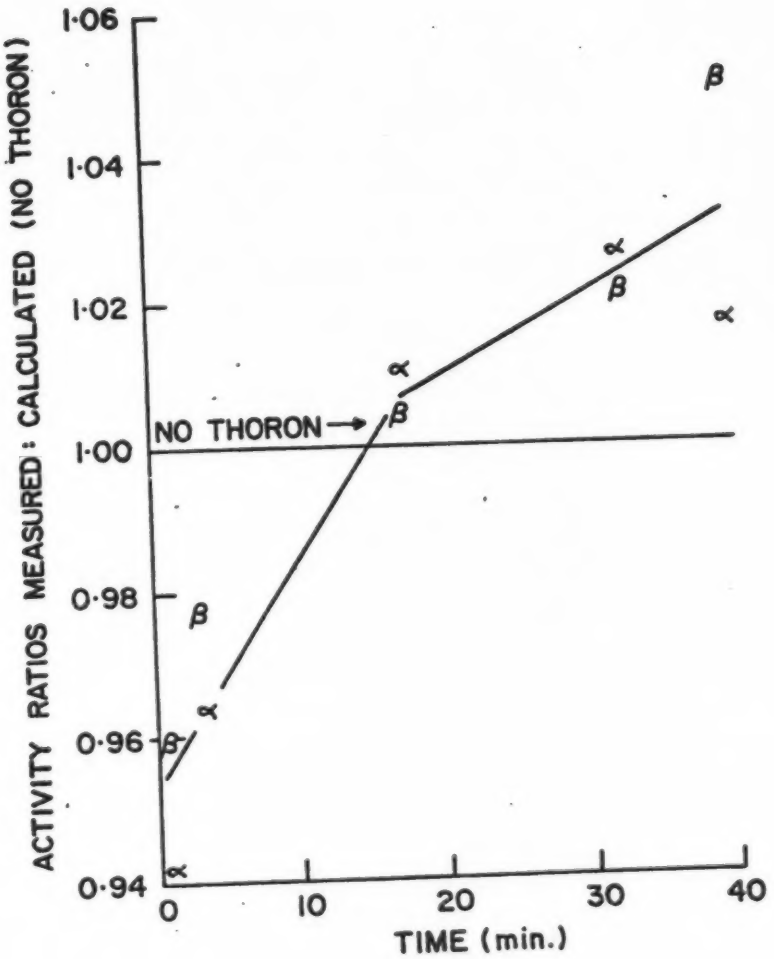

Figure 13. Sample #4, Table VII, Ratios Measured:Calculated

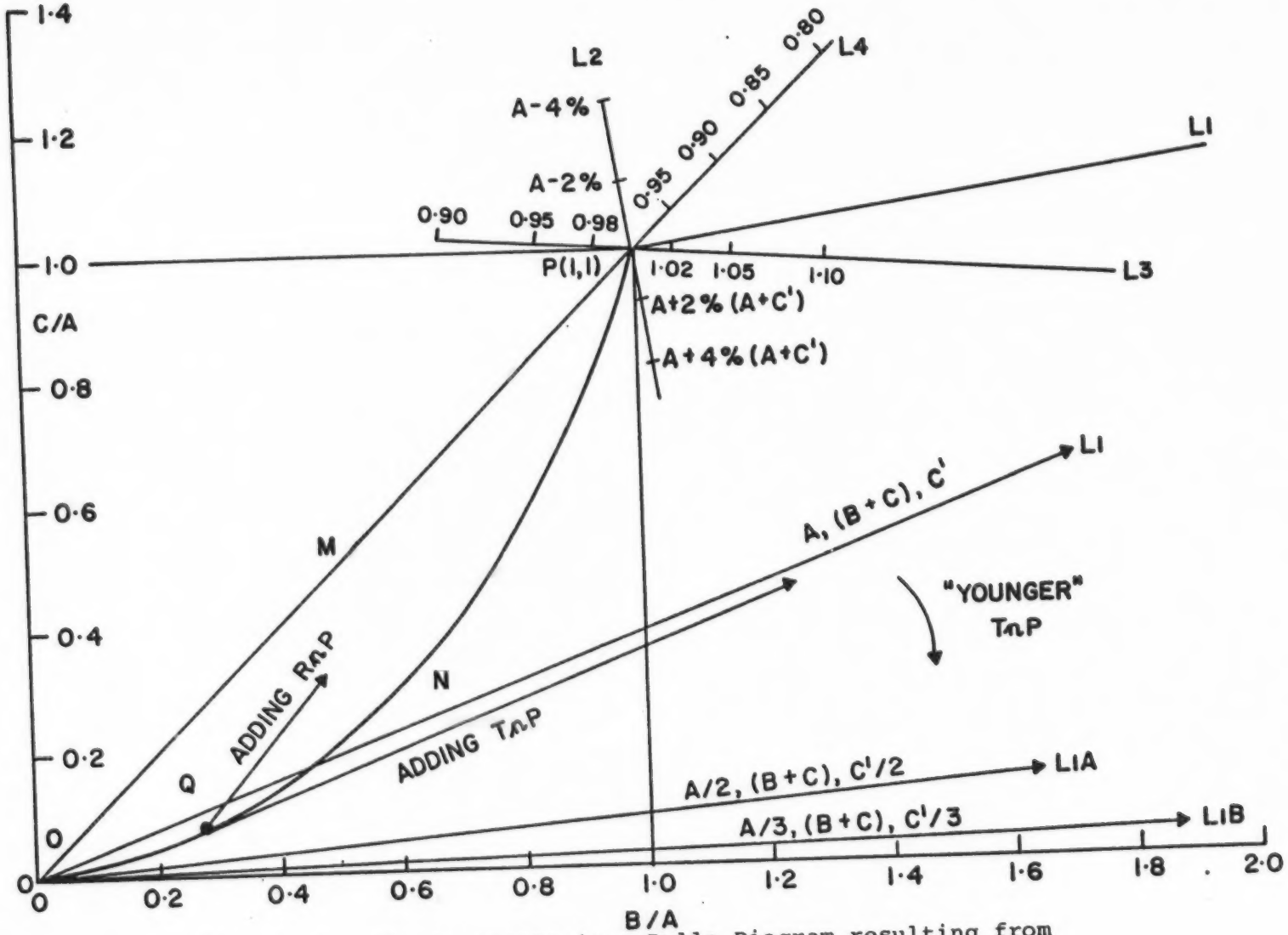

Figure 14. Displacements in a Rolle Diagram resulting from Changes in the A, (B+C), C' Channels, TnP, etc.

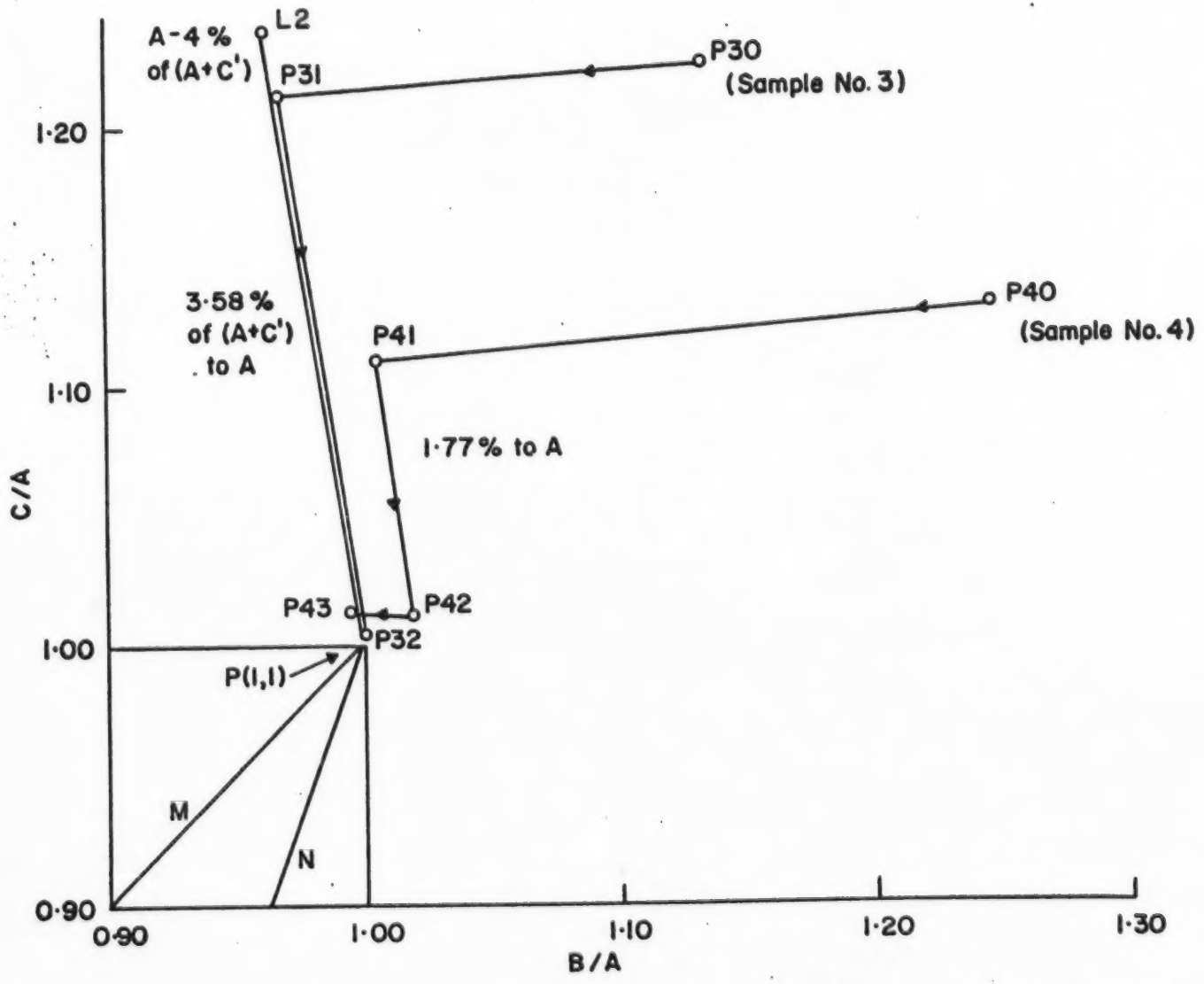

Figure 15. Analysis of Displacements, Samples #3 and #4, Table VII

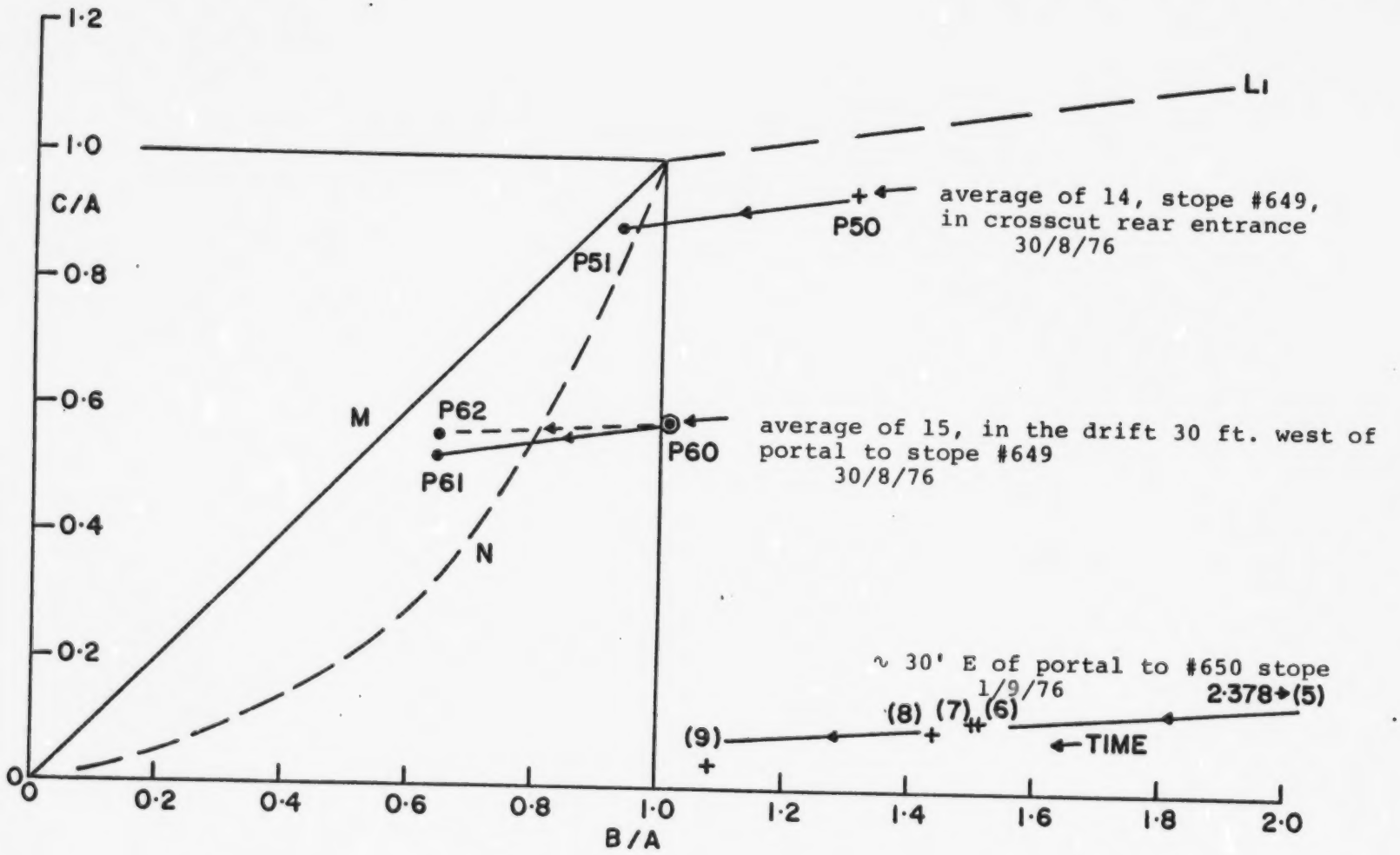

Figure 16. Rolle Diagram of Other Samples Taken Using Argon IWLM